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# Solid State Laser and Second Harmonic Generation

Laboratory Guide to the Lab Work No 4

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#### The aim of the work:

To familiarize with the principle of the solid-state Nd:YAG laser and the effect of second-harmonic generation.

#### **Preleminary task**

- 1. To study the principle of operation and the method of creating an inversion in the Nd:YAG laser.
- 2. Calculate the phase-matching angle when the frequency of the Nd:YAG laser radiation doubles in a KDP crystal, if it is known that for KDP  $n_0$  (1.06  $\mu$ m) = 1.507,  $n_0$  (532 nm) = 1.528, and  $n_e$  (532 nm) = 1.482.

### Theory

The term "solid-state laser" refers primarily to lasers whose active centers are impurity ions introduced into a transparent matrix (crystal or glass). Semiconductor lasers, as they have other pumping and generation mechanisms, are usually classified as a separate class of lasers.

Impurity ions, which are used in solid-state lasers, pertain to one of the groups of transition elements of the periodic table, especially for rare-earth or transition-metal ions. As the matrix crystals, either oxides, for example Al203, or fluorides, for example YLiF4 (YLF) are used. The Al3 + node of the crystal lattice is very small so that the rare-earth element ion can be accommodated in it, and this assembly is mainly used for transition-metal ions. To obtain synthetic garnets, such as Y3Al5O12 = (1/2) (3Y2O3 + 5Al2O3), appropriate combinations of oxides are often used, in which case the Al3 + node can accommodate transition metal ions, whereas the Y3 + node can be used For ions of rare-earth elements. Other oxides include YVO4 crystal for Nd3 + ions and alexandrit for Cr3 + ions. Among the fluorides, YLF material is used as the matrix crystal for ions of rare-earth elements, whereas the most popular materials for transition elements (mainly for Cr3 + ions) are LiSrAlF6 (abbreviated LiSAF) or LiCaAlF6 (succinctly LiCAF).

Comparing oxides and fluorides, it can be noted that the former, being more solid, have some advantages. In particular, they are more preferable in terms of mechanical and termomechanical properties (for example, a higher temperature threshold for destruction). On the other hand, fluorides have better thermo-optic properties (for example, less pronounced induced thermal lenses or induced birefringence).

Among the solid-state lasers, the most known are the ruby laser (the first operating laser), neodymium lasers, a titanium-sapphire laser, erbium and ytterbium lasers. Recently, fiber and disk lasers have developed a great deal, especially in the part of obtaining high power in the infrared range.

### Nd:YAG Laser

The active medium in Nd:YAG lasers is the  $Y_3A1_5O_{12}$  crystal (commonly referred to as YAG or aluminum-yttrium garnet), in which part of the  $Y^{3+}$  ions is replaced by Nd<sup>3+</sup> ions. Typical levels of doping for an Nd:YAG crystal are about 1 at. %. Higher doping levels lead to quenching of fluorescence, as well as to internal stresses in the crystal, since the radius of the Nd<sup>3+</sup> ion is approximately 14% higher than the radius of the Y<sup>3+</sup> ion. Unalloyed starting materials are usually transparent, and after alloying, the YAG crystal acquires a pale purple color, since the absorption lines of Nd<sup>3+</sup> lie in the red region.

Pumping of the active medium of the laser is carried out with the aid of laser diodes with a wavelength of radiation falling into the absorption band of the YAG crystal. In Fig. 1 presents a simplified diagram of the energy levels of the crystal Nd:YAG. The two main pump bands for Nd:YAG correspond to wavelengths of 730 and 800 nm, although other, higher lying absorption

bands (Fig. 2) also play an important role, especially when using flash pump lamps. The absorption bands are associated by fast nonradiative relaxation with a  ${}^{4}F_{3/2}$  level, from which relaxation occurs due to radiation to lower levels ( ${}^{4}I_{9/2}$ ,  ${}^{4}I_{11/2}$ ,  ${}^{4}I_{13/2}$ , etc.). However, the speed of such a relaxation is much less, since the transition in an isolated ion is forbidden, but becomes weakly resolved due to interaction with the crystal lattice field. Nonradiative relaxation in this case is not significant due to screening of both states  ${}^{5}s_{2}$  and  ${}^{5}p_{6}$ , as well as a large energy gap between the  ${}^{4}F_{3/2}$  level and the lower level nearby. Thus, the  ${}^{4}F_{3/2}$  level stores a large fraction of the pump energy and is therefore well suited for the role of the upper laser level. The lifetime of this level is 230 µs.



Fig. 1. Scheme of the energy levels of an Nd: YAG crystal



Fig. 2. Room-temperature absorption spectra of 2.0%Nd:CaF<sub>2</sub>, 2.0%Nd, 2.0%Y:CaF<sub>2</sub> and 2.0%Nd, 6.0%Y:CaF<sub>2</sub> crystals.

Of the possible transitions from the  ${}^{4}F_{3/2}$  level to the underlying I levels, the most intense is the  ${}^{4}F_{3/2} \rightarrow {}^{4}I_{11/2}$  transition. In addition, the  ${}^{4}I_{11/2}$  level is fast (of the order of nanoseconds)

nonradiative relaxation to the ground state  ${}^{4}I_{9/2}$ , so the thermal equilibrium between these two levels is established very quickly. Thus, laser generation at the  ${}^{4}F_{3/2} \rightarrow {}^{4}I_{11/2}$  transition corresponds to a four-level scheme. The  ${}^{4}F_{3/2}$  level is split by the Stark effect into two sublevels (R<sub>1</sub> and R<sub>2</sub> in Fig. 1), while the  ${}^{4}I_{11/2}$  level is split into six sublevels. Laser generation usually occurs from the upper sublevel R<sub>2</sub> to a specific sublevel of the  ${}^{4}I_{11/2}$  level, since this transition has the largest cross section for the transition of stimulated emission. This transition is carried out at a wavelength  $\lambda = 1.064 \mu m$ , the most common wavelength of generation for Nd:YAG lasers. Laser generation can also be obtained at the  ${}^{4}F_{3/2} \rightarrow {}^{4}I_{13/2}$  transition with a wavelength  $\lambda =$ 1.319 µm, realizing a multilayer dielectric coating on the mirrors of the resonator. When laser diodes are used as a pump, laser generation can also be efficiently performed at the transition  ${}^{4}F_{3/2} \rightarrow {}^{4}I_{9/2}$  ( $\lambda = 946$  nm).

Nd:YAG lasers have a rather narrow spectral line -  $\Delta v = 4.2 \text{ cm}^{-1} = 126 \text{ GHz}$  at room temperature. Most often, the active medium is made in the form of a rod, the diameter of which is usually from 3 to 6 mm and a length of 5 to 15 cm. They can work in both continuous and pulsed modes, while pumping can be carried out as a lamp , And semiconductor AlGaAs laser. As a source of radiation under lamp pumping, medium-pressure xenon lamps are used for pulsed operation and high-pressure krypton lamps for non-continuous pumping. As a rule, linear lamps with a close arrangement of the lamp and crystal are used. In both pulsed and continuous modes, the differential efficiency of the laser (the ratio of the laser generation energy to the absorbed pump energy) during lamination pumping is about 3%, the average output power reaches several kilowatts.

When working with laser diode pumping, the radiation is introduced either from the end of the crystal along the laser generation axis (longitudinal pumping, Fig. 3a) or along the lateral surface of the crystal (transverse pumping, Fig. 3b). To ensure high pumping power and convenience of introducing radiation into the crystal, diode lasers conjugated with optical fibers are often used. Continuous Nd:YAG lasers with longitudinal pumping by laser diodes provide an output power of up to 15 W. In the case of transverse pumping, the output power of such lasers today reaches 100 W and higher. Differential efficiency when using diode pumping is much higher compared with a lamp and can exceed 10%.



Fig. 3. Schemes of longitudinal (a) and transverse (b) pumping of an Nd: YAG laser

Nd:YAG lasers are widely used in various fields of science and technology, in particular:

1. Processing of materials (drilling, welding, etc.).

2. Application in laser range-finding for military tasks, especially for laser viewfinders and target pointers.

3. In medicine, it is used for coagulation and for the cutting of tissues, for the destruction of unnoticed membranes of pathological formations (for example, secondary cataracts), for the treatment of iritectomy, etc.

4. Scientific research.

5. Generation of the second harmonic in nonlinear crystals. Nd: YAG lasers with diode pumping and a built-in intracavity device for second-harmonic generation allow the continuous output of radiation with a wavelength of  $\lambda = 532$  nm and a power of up to ~10 W, and are a worthy alternative to Ar A laser in many applications

## In-lab task

- 1. To familiarize yourself with the principle of operation and functional designation of the IMO-2 and Ophir PD-300 radiation power meters with a display.
- 2. To study the design, principle and operating conditions of a helium-neon laser (for example, the lasers GN-3-1 and LH-56).
- 3. Visually determine the wavelength of the radiation.
- 4. Measure the output power of the laser radiation (using the IMO-2 and Ophir PD-300 receivers).
- 5. Estimate the full efficiency of the laser, if the power consumption of the network is 15 watts.
- 6. Express your thoughts on the degree of danger of working with laser radiation.
- 7. Using a collecting lens, measure the power of the incandescent lamp (measurements are made using IMO-2).
- 8. Explain the difference between lasers and sources of spontaneous emission.

## Second harmonic generation

In classical linear optics, it is assumed that the induced electrical polarization of the medium depends linearly on the applied electric field, i.e.

 $P = \varepsilon_0 \chi E$ , (2.1)

where  $\chi$  is the linear dielectric susceptibility of the medium. For strong electric fields characteristic of laser beams, the relation (2.1) is no longer a good approximation, and it is necessary to take into account the subsequent expansion expansions in which the vectors P should be regarded as functions of higher Powers of E, for example:

$$\mathbf{P} = \varepsilon_0 \chi \mathbf{E} (1 + \alpha_1 \mathbf{E}), (2.2)$$

where the coefficient  $\alpha 1$  describes the nonlinear dependence of the polarization P on E. With a nonlinear term of polarization

 $P^{\rm NL} = \epsilon_0 \chi \alpha_1 E^2 \ (2.3)$ 

two main effects are connected:

1. Generation of the second harmonic (SHG), at which a laser beam with frequency  $\omega$  is partially transformed by a nonlinear medium into a coherent beam with a frequency  $2\omega$ .

2. Optical parametric generation (OPG), in which a laser beam with a frequency  $\omega_3$  causes a simultaneous radiation of two coherent beams with frequencies  $\omega_1$  and  $\omega_2$  in a nonlinear material, with  $\omega_1 + \omega_2 = \omega_3$ .

For strong electric fields that occur in laser beams, the conversion efficiency in both these processes can be very high (approaching 100% in the case of SHG). Therefore, at present these methods are used to generate new coherent waves with different frequencies, which differ from the frequency of the incident wave.

To perform this laboratory work we use the Nd:YAG laser with frequency conversion, which is based on the SHG effect. Therefore, let us dwell in more detail on this effect.

Consider a monochromatic plane wave with frequency  $\omega$  propagating in the z direction through a nonlinear crystal. In this case, it is assumed that the origin of the z-axis coincides with the input end of the crystal. For the electric field  $E_{\omega}(z, t)$  of a plane electromagnetic wave, we can write the following expression:

$$E_{\omega}(z,t) = E(z,\omega) \exp\left[i\left(\omega t - k_{\omega}z\right)\right], (2.4)$$

where  $k_{\omega} = 2\pi/\lambda = n_{\omega}\omega/c$  is the wave number at the fundamental frequency,  $n_{\omega}$  is the refractive index at frequency  $\omega$ , and c is the speed of light in vacuum.

In accordance with (2.3), the light wave propagating in the medium causes a wave of nonlinear polarization of the crystal

$$P_{2\omega}^{\rm NL} = \varepsilon_0 \chi \alpha_1 E^2(z,\omega) \exp\left[i\left(2\omega t - 2k_{\omega}z\right)\right]. (2.5)$$

This expression describes a polarization oscillating at a frequency  $2\omega$  and propagating in space in the form of a wave with wave number  $2k\omega$ . The electric field of this electromagnetic wave is written in the form:

$$\mathbf{E}_{2\omega}(\mathbf{z},\mathbf{t}) = \mathbf{E}(\mathbf{z},2\omega) \exp\left[i\left(2\omega\mathbf{t}-\mathbf{k}_{2\omega}\mathbf{z}\right)\right], (2.6)$$

where  $k_{2\omega} = 2n_{2\omega}\omega/c$  is the wave number at the second harmonic frequency,  $n_{2\omega}$  is the refractive index at the frequency  $2\omega$ .

The physical meaning of SHG can be understood as the result of the beating of an electromagnetic wave at the fundamental frequency  $\omega$  with itself, which leads to a polarization oscillating with a frequency of  $2\omega$ . From expressions (2.5) and (2.6), one can obtain a condition that must be satisfied so that the SHG process proceeds effectively. Namely, the phase velocity of the polarization wave (vP =  $2\omega/2k_{\omega}$ ) should be equal to the phase velocity of the generated electromagnetic wave (vE =  $2\omega/k_{\omega}$ ). This condition can be written in the form:

$$k_{2\omega} = 2k_{\omega} . (2.7)$$

If this condition is not satisfied, then at some distance l inside the crystal the phase of the polarization wave  $(2k_{\omega}l)$  will differ from the phase  $(k_{2\omega}l)$  of the generated wave, which comes from the point z = 0 to the point z = 1. This phase difference  $(2k_{\omega}-k_{2\omega})l$  increasing with distance l means that the generated wave will not grow cumulatively with distance l, since it is not supported by polarization with the corresponding phase. Therefore, condition (2.7) is called the condition of phase synchronism. It can also be written in terms of the refractive index of the crystal:

 $n_{2\omega} = 2n_{\omega}$ 

Let us further consider the features of wave propagation in anisotropic crystals, which are used for SHG. In such crystals, two different linearly polarized plane waves with different phase velocities can propagate in a given direction. Two different refractive indices correspond to these two different polarizations. Such a difference in the values of the refractive indices is called birefringence. To describe this phenomenon, a so-called ellipsoid of refractive index is usually used, which in the case of a uniaxial crystal is an ellipsoid of revolution around the optical axis (z-axis in Fig. 4a). The two allowed directions of polarization and the corresponding refractive indices are determined as follows. Through the center of the ellipsoid, a straight line is made in the direction of propagation of the beam (the straight line OP in Fig. 4a) and the plane perpendicular to this line. The intersection of this plane with an ellipsoid-house forms an ellipse. The two axes of the ellipse are parallel to the two directions of polarization, and the length of each half-axis is equal to the value of the refractive index for a given direction of polarization. One of these directions is necessarily perpendicular to the optical axis, and a wave having such a direction of polarization is called ordinary-wave. It can be seen from the figure that its refractive index n<sub>0</sub> does not depend on the direction of propagation. A wave with a different polarization direction is called an extraordinary wave, and the value of the corresponding refractive index  $n_e(\theta)$  depends on the angle  $\theta$  and varies from the refractive index of the ordinary wave  $n_0$  (when the OP is parallel to the z axis) N, called the refractive index of an extraordinary wave (when the OP is perpendicular to the z axis). A positive uniaxial crystal corresponds to the case  $n_e > n_0$  (Fig. 4a), and a negative uniaxial crystal -  $n_e < n_0$  (Fig. 4b).



Fig. 4. The ellipsoid of the refractive indices (a), the phase-matching angle  $\theta$ m in the case of second-harmonic generation (b)

To satisfy the condition of phase synchronism, the basic wave can be started at an angle  $\theta$ m to the optical axis so that

$$n_{e}(2\omega,\theta_{m}) = n_{0}(\omega) \cdot (2.9)$$

Fig. 4b shows the intersections of the surfaces of the normals  $n_0(\omega)$  and  $n_e(2\omega, \theta)$  with the plane containing the z axis and the direction of propagation. Because of the normal dispersion of the crystal,  $n_0(\omega) < n_0(2\omega)$  does not hold, whereas for a negative one-axis crystal we have  $n_e(2\omega) < n_0(2\omega)$ . According to Fig. 4, one can write  $n_e(2\omega) = n_e(2\omega, 90^\circ)$  and  $n_0(2\omega) = n_e(2\omega, 0)$ . It

follows that the "ordinary" circle for frequency  $\omega$  intersects the "extraordinary" ellipse for the frequency  $2\omega$  for some value of the angle  $\theta_m$ . For all rays lying on the surface of the cone of rotation around the z axis with an angle  $\theta_m$  at the vertex, condition (2.9) is satisfied and, consequently, the phase matching condition is satisfied.

If we enter the Cartesian coordinates z and y for an arbitrary point of the ellipse describing the refractive index  $n_e(2\omega, \theta)$  of the extraordinary wave, then we can write:

$$\frac{z^2}{[n_0(2\omega)]^2} + \frac{y^2}{[n_e(2\omega)]} = 1. (2.10)$$

If the coordinates of z and y are expressed in terms of the quantity  $n_e(2\omega, \theta)$  and the angle  $\theta$  is  $\omega$ -responsible, then (2.10) takes the form:

$$\frac{\left[n_{e}(2\omega,\theta)\right]^{2}}{\left[n_{0}(2\omega)\right]^{2}}\cos^{2}\theta + \frac{\left[n_{e}(2\omega,\theta)\right]^{2}}{\left[n_{e}(2\omega)\right]}\sin^{2}\theta = 1. (2.11)$$

For  $\theta = \theta_m$  and substituting the expression (2.9) into (2.11), we obtain the following:

$$\left[\frac{n_0(\omega)}{n_0(2\omega)}\right]^2 (1-\sin^2\theta_m) + \left[\frac{n_0(\omega)}{n_e(2\omega)}\right]^2 \sin^2\theta_m = 1. (2.12)$$

Solving the last equation, for the variable  $\sin 2\theta_m$  we obtain the expression for the phasematching angle:

$$\sin^2 \theta_{\rm m} = \left( \left[ \frac{n_0(2\omega)}{n_0(\omega)} \right]^2 - 1 \right) / \left( \left[ \frac{n_0(2\omega)}{n_e(2\omega)} \right]^2 - 1 \right). (2.13)$$

We note that under certain conditions (for example, when the temperature of the crystal changes), the spherical surface of the refractive index for the ordinary fundamental wave does not intersect the ellipsoidal surface of the second harmonic of the extraordinary wave, but only touches it. Then the angle  $\theta$ m is 90 ° and the phase synchronization becomes much less critical (in terms of angle). This type of phase synchronism is called ninety degree phase synchronism.

Nonlinear crystals, most often used as doublers of the radiation frequency of an Nd: YAG laser, include KTP crystals (potassium titanyl phosphate) and BBO (barium beta-borate). KDP crystals (potassium dihydrogenphosphate), DKDP (potassium dideoxyphosphate), ADP (ammonium phosphate), etc. are also used.

#### Functional scheme of Nd: YAG laser with second harmonic generation

The functional scheme of the Nd: YAG laser considered in this laboratory work is shown in Fig. 5. Intracavity transformation occurs in the laser into the second harmonic of radiation with a wavelength  $\lambda = 1.06 \mu m$ . The radiation from the array of laser diode through the matching optics is focused on the Nd:YAG rod. As a frequency converter, a nonlinear crystal KTP (KTiOPO4), which does not require temperature control, is used. The frequency converter includes a special output mirror ("dull" at the fundamental wavelength  $\lambda = 1.06 \mu m$  and transmitting at a wavelength of the harmonic  $\lambda = 532 \text{ nm}$ ), a nonlinear crystal and an optical filter. The optical filter is the third intracavity mirror designed for unidirectional output of second-harmonic radiation. The emitter and the converter are made in the form of separate blocks and are adjusted directly in the laser casing. BP - pulse power supply for laser diodes.



Fig. 5. Functional scheme of Nd: YAG laser with second harmonic generation

## In-Lab task

1. To study the functional scheme and principle of operation of a neodymium laser.

2. Measure the output power of the laser radiation of laboratory lasers LT-01 and LS-1-LN-532-200 using the Ophir PD-300 power meter.

3. Visually determine the wavelength of the radiation.

4. Using the photodiode DET10A and the oscilloscope, measure the pulse length of the generation pulse and the repetition rate of the pulses of LT-01 laser.

5. Calculate the energy in the generation pulse.

## **Control questions**

1. Explain the principle of inversion formation in the four-level scheme and its difference from the three-level scheme.

- 2. What is the quantum efficiency of the Nd:YAG laser?
- 3. In what modes can the Nd:YAG laser work?
- 4. What wavelengths can be obtained from an Nd:YAG laser?
- 5. What type of pumping is used in Nd:YAG lasers?
- 6. In which regions of the spectrum are the main absorption bands of the  $Nd^{3+}$  ion?
- 7. What is the second harmonic generation effect used for?
- 8. Why should the phase-matching condition be satisfied?
- 9. What are the main types of nonlinear crystals used for SHG?
- 10. What is the difference between a positive and negative uniaxial crystal?
- 11. Explain the differences between the effects of SHG and OCG.