doi: 10.56761/EFRE2022.N4-O-011202

Superluminescence of NV centers in diamond pumped by the second harmonic of a Nd:YAG laser

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Abstract. In the light of the creation of a diamond laser [1], an urgent task is to determine the characteristics of diamonds in order to determine the range in which such lasers can be created not in isolated cases. This work is aimed at studying the mechanisms of creating superluminescence in diamond under the action of optical pumping by the second harmonic of an ND:YAG laser (λ = 532 nm).

It was found that, upon pulsed excitation of a diamond by an ND:YAG laser with a pump intensity above ~2.0 MW/cm^2 in the spectral range 700–750 nm, a nonlinear increase in intensity appears against the background of the spontaneous photoluminescence spectrum, which, with a further increase in the pump intensity, turns into a pronounced superluminescence peak with a maximum at about 718 nm. An increase in the pump intensity from 2.7 to 46 MW/cm^2 leads to the broadening of this peak at half maximum from 13 to 19 nm. At high levels of pump intensity, nonlinear absorption of pump radiation and accumulation of NV centers in the excited state were found.

The position of the photoluminescence band was calculated as a function of various values of the inversion density of the populations of color centers, taking into account the intrinsic absorption spectrum of diamond. The calculation results are close to the experimental data.

Keywords: diamond, laser, optical amplification, superluminescence, NV center.

1. Introduction

The creation of optical sources with high thermal conductivity, chemical and radiation resistance, and mechanical strength has been and remains an urgent task. Such light sources are in demand in many areas, and with the development of science and technology, this need is only increasing. As separate examples, it can be mentioned that powerful LEDs with high thermal conductivity are needed for street lighting, LEDs, semiconductor lasers with high radiation resistance are needed in the nuclear and space industries. There is a tendency to switch from electrical signals that control equipment to optical ones, and this requires light sources that work stably in "close to combat" conditions.

The most suitable for the requirements of developing technology is diamond. In this connection, there is now a rapid growth in both the number of crystal synthesis plants and the development of synthesis methods in order to create diamonds with desired characteristics. To create light sources based on diamond, the necessary changes must be made to the crystal during its synthesis, impurities are added, and defects are created [2–6]. The results of studies of defects (color centers) with the aim of creating, for example, a laser active medium have been published in many papers. An analysis of these publications [7–10] shows that diamonds with NV centers, i.e., color centers based on nitrogen atoms embedded in the diamond lattice, look very promising for solving this problem. These centers have broad absorption and luminescence bands, a high gain, and a short lifetime at the excited level, which indicates the possibility of creating laser-active media on their basis.

In this paper, we present a continuation of studies [11] of superluminescence under the action of second harmonic radiation from an ND:YAG laser, namely, the effect of absorption on the characteristics of superluminescence in diamond.

2. Experiment description and results

2.1. Experimental setup

In this work, we continued the study of a diamond sample synthesized by the temperature gradient method (high pressure, high temperature (HPHT). A 4.4 × 4.4 × 0.25 mm plate was cut from the grown diamond along the electron beam irradiation and subsequent annealing created color centers (Fig.1).

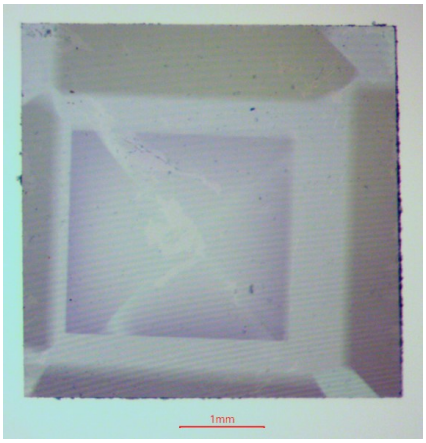

Fig.1. Sample photo. The numbers show the zones of interest with color centers.

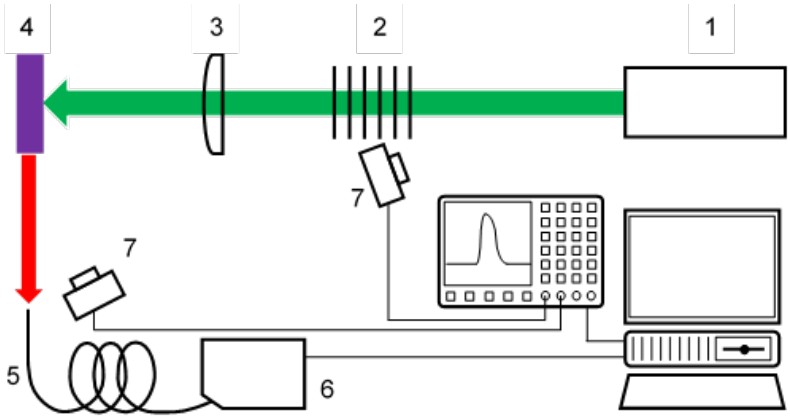

Fig.2 Scheme of the experimental setup. 1 – Nd:YAG laser; 2 – light filters; 3 – cylindrical lens; 4 – diamond sample; 5 – light guide; 6 – spectrometer; 7 – avalanche photodiodes

Experiments on obtaining superluminescence were carried out on a setup, the scheme of which is shown in Fig.2. Pulsed radiation with a wavelength of 532 nm (the second harmonic of the Nd:YAG laser (1) was used to excite the luminescence of the sample. The pump intensity was in the range of 0.1–50 MW/ cm^2 was varied by neutral filters (2). The pumping radiation was focused by a cylindrical lens (3) into one of the zones (Fig.1) of the sample (4). The luminescence of the sample was recorded at the end of the plate at an angle of 10–30 degrees to the plane of the plate (direction 100). The luminescence yield in this direction was maximum. Luminescence emission from the light guide (5) in the range of 200–900 nm was fed into the AvaSpec-2048-2 spectrometer (6). The spectral data, taking into account the spectral sensitivity of the device and the transmission of the optical fiber, were accumulated in a digital file. pump pulse, its duration, as well as the energy and duration of the luminescence pulse were recorded by an Ophir sensor and an avalanche fotodiode (7). The pump energy per pulse was 80 mJ for a pulse duration of about 10 ns at half maximum.

From the transmission spectra, the absorption spectra and changes in the absorption index were calculated. Knowing the index β and the absorption cross section σ (taken from the literature ($\sigma = 2.8\ 10^{-17}$ cm^2), the concentration of color centers N in the corresponding zones was calculated using formula (1):

$$N = \beta/\sigma. \quad (1)$$

Table 1 shows the results of the analysis of the spectra of zones with created defects.

Table 1. Concentration of substitutional nitrogen and NV- centers in the zones

Zone	N_c, $\times 10^{19}$ cm^{-3}	N_{NV^-}, $\times 10^{19}$ cm^{-3}	β_{NV^-}, cm^{-1}
1	2.55	0.066	18.5
2	0.085	0.018	5.0
3	2.21	0.064	18.0

It can be seen from the table that the central part of the crystal (zone 2) is a region with a low content of the substituting nitrogen N_c, therefore, in this zone, the luminescence intensity is weak, that is, this region is unsuitable for studying the required centers, and is not considered further. Zones 1 and 3 have similar spectra, consisting of an intense absorption band by NV^- centers, with an exponent of ~18 cm^{-1}. It is in these zones that superluminescence was obtained

2.2. Experimental setup

The photoluminescence spectra of the sample depending on the pumping intensity are shown in Fig.3. The spectra are normalized to the intensity of the maximum of the unperturbed phonon wing of NV^- centers at 680 nm.

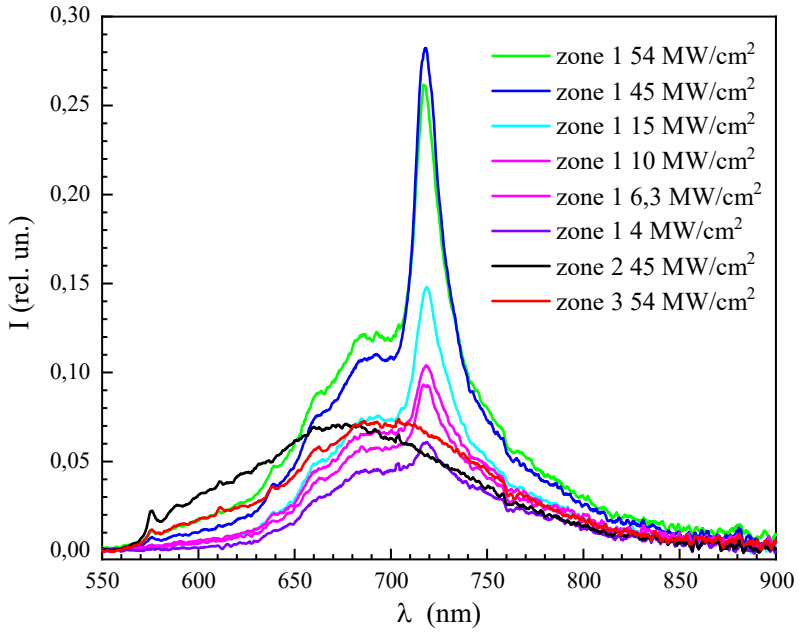

Fig.3. Photoluminescence spectra, at different pumping intensities, in different zones.

In the photoluminescence spectra, there is a noticeable band of NV^- centers with a distinguishable zero-phonon line (ZPL) (λ = 638 nm). As the pump intensity increases above ~2.0 MW/cm^2, a nonlinear increase in intensity is observed in the spectral region 700–760 nm, and at a pump intensity above 2.5 MW/cm^2 this band turns into an intense peak with a maximum of

~718 nm. The half-width of this peak increased from 13 to 19 nm as the pump intensity increased from 2.7 to 46 MW/cm^2.

3. Discussion of results

First, we note an important point regarding the method of gain calculations and terminology. As a criterion for the suitability of using color centers as work centers, a high radiative transition cross section is used. The cross section of the radiative transition is usually calculated for the maximum of the luminescence band, and lasing is expected to be obtained in the same band. In many works (including ours), the radiative transition cross section (gain cross section) σ multiplied by the number of centers in the excited state N^* is identified with the gain index:

$$\alpha = N^* \cdot \sigma. \quad (2)$$

In this case, the losses are assumed to be spectrally independent. A high cross section of the radiative transition in color centers is a necessary but not sufficient condition for obtaining amplification and generation of laser radiation. According to this calculation, for example, the fact that the ground electronic state of the center can be populated, and, consequently, amplification and generation in the ZPL region, is not taken into account is possible only if more than half of the centers are excited. In addition, the characteristics of the crystal – the matrix in which the color centers are contained – are usually not taken into account. There are losses in the crystal, which are due both to self-absorption in the vicinity without a phonon line, and a number of other losses not related to color centers. Such losses are, as a rule, spectrally dependent and constitute the absorption spectrum of the crystal.

In the absorption spectrum of our synthetic diamond (Fig.4), the NV^- color centers appear as an intense band with a maximum of ~650 nm. In addition, the absorption spectrum contains an absorption "step" near the ZPL centers and a band with a maximum of ~785 nm, which is a source of additional losses that adversely affect lasing.

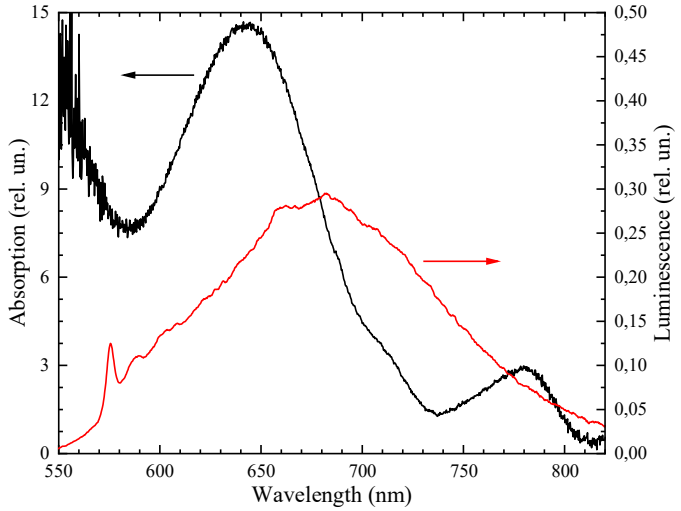

Fig. 4. Absorption and luminescence spectra of a crystal containing NV^- centers.

As follows from Fig.4, the luminescence maximum falls on the ~680 nm region of the spectrum, which corresponds to the radiative transition cross section of $4.3 \cdot 10^{-17}$ cm^2. It is in this region of the spectrum that the appearance of superluminescence should be expected. However, both in [11] and here, generation or superluminescence is found in the region of ~720 nm (Fig. 3).

As mentioned above, when propagating in a medium with population inversion, the radiation will be amplified with an amplification factor α(λ) proportional to the concentration of excited centers N^* and the cross section of the radiative transition $\sigma_i(\lambda)$ (formula (2)). At the same time, in a

diamond sample, there are losses due to absorption, scattering, reflection from faces, etc. These losses are also spectrally dependent and together make up the absorption spectrum of the crystal β(λ) (formula (1)) (Fig.4). Therefore, the propagation of radiation in an amplifying medium at a wavelength λ with spectrally dependent losses can be written as:

$$I(\lambda) = I_0 \exp[\alpha(\lambda)-\beta(\lambda)]d, \quad (3)$$

In relation (1), β(λ) in this case is an indicator of the total optical loss at wavelength λ. From (3) the threshold population inversion density is determined, the excess of which will lead to amplification. The threshold density of population inversion is defined as N^* for which $I(\lambda)/I_0 > 1$ or

$$I(\lambda)/I_0 = \exp[N^* \sigma(\lambda)-\beta(\lambda)]d > 1, \quad (4)$$

or

$$N^* \sigma(\lambda)>\beta(\lambda), \quad (5)$$

That is, the intensity of radiation propagating in diamond will increase at those wavelengths for which the gain exceeds the losses. In our case, this condition is satisfied by the range of 700–730 nm, where the luminescence is quite intense, and absorption losses are minimal (Fig.4). With a further increase in the inversion density, the radiation intensity increases, and the band in which the generation frequency can be tuned expands. Note that in the region of a zero-phonon line, generation of laser radiation is impossible, although the calculation of the cross section for the transition from the upper laser level to the lower ones gives non-zero values.

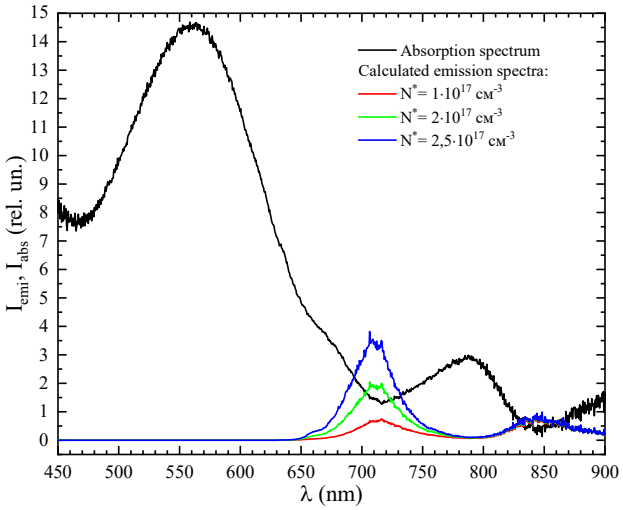

Fig. 5. Absorption and luminescence spectra of a crystal with NV centers.

Thus, the use of the intrinsic absorption spectrum takes into account the fact that the ground electronic state of the color center is populated and there is no population inversion with respect to this state. In addition, the position of the superluminescence band is determined not by the maximum in the luminescence spectrum, but by the maximum of the loss ratio. Substituting into formula (4) the real values for zone 1 of our sample at the population inversion density $N^* = 10^{17}$ cm^{-3}, we find that the gain will be 1.47 (times). This means that the radiation from the narrow edge of the sample will be amplified (in one pass) by approximately one and a half times, which, in the presence of a tuned resonator, is sufficient to obtain stable lasing.

Fig.5 shows the calculated emission spectra calculated according to (4) at population inversion densities $N^* = 1\cdot10^{17}$, $2\cdot10^{17}$, and $2.5\cdot10^{17}$ cm^{-3} and medium length $d = 1$ cm. Comparing Figs.5 and 3, we find a correspondence between the calculated and experimental results.

4. Conclusion

In connection with the obtained research results, the following conclusions can be drawn. The most probable reason for the decrease in the luminescence intensity with increasing pump power is the absorption of the second photon by color centers in the excited state and the transition of the centers to higher metastable levels.

The observed nonlinearities in the absorption of optical pump energy and the concentration of color centers in the excited state require additional studies of the physics of these processes. In particular, calculations of possible radiation amplification in further studies should be carried out taking into account the intrinsic absorption spectrum of the sample. At high pump intensity levels, the effect of multiphoton and multistage absorption processes should be studied. Also, it is necessary to study various types of luminescence quenching, for example, concentration quenching, temperature quenching, quenching by nitrogen and other impurity defects, etc. Absorption losses in the region of laser radiation generation should be minimized. Which, in turn, implies increased requirements, if not for the creation of synthetic diamonds at this stage of technology development, then at least for the selection of samples with identical characteristics.

Acknowledgements

The study was carried out on the state order of the Ministry of Science and Higher Education of the Russian Federation, project № FSWM-2020-0048.

5. References

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