

PACS 78.55.-m

Effect of non-linear absorption on characteristics of laser-induced luminescence

S.E. Zelensky, B.A. Okhrimenko

*Taras Shevchenko Kyiv National University, Physics Department,
6, prospect Academician Glushkov, 03680 Kyiv, Ukraine;
E-mail: zele@univ.kiev.ua*

Abstract. This paper deals with the methodology of laser-induced luminescence at high excitation levels. For a case when a photodetector collects integral (over the volume) luminescence power, a method is proposed for processing the experimentally-measured luminescence power and optical transmittance as some functions of the incident laser power. The method is based on the analysis of relative increments of luminescence and transmitted laser power. Considered are two examples of mechanisms of non-linear laser-induced luminescence, namely: saturated molecular luminescence and luminescence excited via two-photon absorption.

Keywords: luminescence, absorption, non-linearity, laser induction.

Manuscript received 14.02.05; accepted for publication 18.05.05.

1. Introduction

Laser-induced luminescence is a powerful tool for investigations of physical properties of molecules and impurity centers in various matrixes. The possibilities of this method are enhanced significantly, if the laser radiation power is sufficiently high to initialize various non-linear processes in the studied object. For example, such processes may be a saturated absorption, spectral hole burning, multiphoton absorption, excited-state absorption, *etc.*

When investigating the laser-induced non-linear processes, the choice of a suitable parameter for characterization of the investigated process is a critical factor. In the traditional non-linear optics, the formalism of non-linear susceptibility tensors is widely used to characterize coherent phenomena (generation of optical harmonics, four-wave mixing, *etc.*). However, for a number of non-coherent phenomena (saturation, non-linear absorption, *etc.*), the non-linear susceptibility approach seems to be inconvenient as it results in complicated equations with the loss of clearness. Choosing a suitable parameter of non-linearity is also impeded by the fact that the experimentalists usually deal with the integral characteristics of luminescence and absorption, with an essentially non-uniform spatial distribution of the laser power.

Consider a simple example. As is theoretically well substantiated for a two-photon mechanism of excitation of luminescence, it is expedient to approximate the

dependence of the luminescence intensity on the excitation one using the following power function

$$I = \text{const } F^\gamma \quad (1)$$

where $\gamma = 2$ for this instance. Whereas the γ -parameter is a constant, and two-photon absorption is usually a low probable process, the integral (over the volume) luminescence power can be also approximated using the function (1). The introduced parameter γ is a convenient characteristic of the investigated process; it can be easily determined from the experimental data (as a slope of the curve $I(F)$ in a log-log scale) and provides the information on the number of photons engaged in the multiphoton absorption transition. It should be also noted that γ is a dimensionless parameter which can be calculated using the experimental data (I and F) measured in suitable arbitrary units.

It is not a rare case in laser spectroscopy when the laser-induced emission is characterized with a non-linear dependence of its power on the excitation intensity. In many these cases, the attempt to approximate the experimental data the function (1) leads to the conclusion that γ is not a constant but depends on the excitation laser power $\gamma = \gamma(F)$. For example, such behavior is observed in the following cases: the saturated luminescence, the laser-induced incandescence [1, 2], the luminescence excited by sequential absorption in YAG:Nd³⁺ [3], *etc.* In the mentioned examples, γ -parameters possess non-integer values and provide the useful information on the investigated laser-induced non-linear

processes. It should be also noted that the dependence $\gamma(F)$ makes difficulties for theoretical calculations and results in significant errors caused by the non-uniformity of laser power distribution within the sample volume [4].

In this paper, we analyze the effect of non-linear absorption of the excitation laser radiation on the experimentally measured luminescent characteristics. Using the above-mentioned γ -parameters both for the luminescence intensity and for the transmitted laser power, we propose a method for processing the experimental data that provides a possibility to distinguish the absorption and luminescence non-linearities.

2. General considerations

Consider the interaction of a laser beam with light-absorbing centers or molecules in a transparent matrix. Assume the surface power density of laser radiation is uniformly distributed within the cross-section area of the laser beam. Denote the surface density of the laser power (expressed in photons /cm²s) for incident and transmitted laser beams by F_0 and F_d , respectively. Denote the optical transmittance of a layer with the thickness d as $T = F_0/F_d$. Let the molecules absorb laser photons and emit the luminescence ones with a quantum efficiency η and the average photon energy $\hbar\omega$. dF signifies the difference of the surface density of the laser power passing through the layer of the thickness dz . P_L identifies the integral power of luminescence emitted in all directions from the total volume $V = Sd$, where S is the cross-section area of the laser beam. Knowing the number of laser photons absorbed in the elementary volume Sdz is $-SdF$, we obtain the following expression for P_L as a function of F_0

$$P_L(F_0) = -\hbar\omega S \int_{F_0}^{F_d(F_0)} \eta(F) dF. \quad (2)$$

If the quantum efficiency of luminescence is independent of the excitation power density, then (2) can be reduced to the following form

$$P_L(F_0) = \hbar\omega\eta S [F_0 - F_d(F_0)]. \quad (3)$$

For the integral luminescence, the following parameter of non-linearity can be defined as a ratio of relative increments

$$\gamma_L = \frac{dP_L/P_L}{dF_0/F_0}. \quad (4)$$

By a little algebra, the expression (3) yields

$$\gamma_L = \frac{1 - (1 + \gamma_T)T}{1 - T}, \quad (5)$$

where the non-linearity parameters for the optical transmittance γ_T and transmitted laser power density γ_F are defined similarly to (4)

$$\gamma_T = \frac{dT/T}{dF_0/F_0} = \frac{dF_d/F_d}{dF_0/F_0} - 1 = \gamma_F - 1. \quad (6)$$

It follows from the expression (5) that γ -parameters of integral luminescence (4) and transmittance (6) are interrelated. As γ_T and T are functions of F_0 , then γ_L is also a function of F_0 according to (5). For example, in a simple case of power-independent absorption ($T = \text{const}$, $\gamma_T = 0$, $\gamma_F = 1$), we obtain $\gamma_L = 1$, that means an ordinary linear response of luminescence intensity to the variations of the excitation power.

It should be emphasized that the expression (5) is derived without any assumption concerning the mechanism of absorption of laser radiation by the luminescence centers. So, it could be both linear and non-linear absorption, for example, single-photon absorption according to Bouguer's law, multiphoton absorption, saturated absorption, etc. In this sense, the expression (5) seems to be a universal formula.

As seen from the expression (2), the non-linear properties of integral luminescence are determined by several factors. (i) The non-linearity of absorption effects on the luminescence. According to (2), the power of luminescence depends on the upper limit of integration F_d that, in its turn, depends on F_0 . That is why the value γ_L and its dependence on F_0 are determined by the values T and γ_T , and by their behavior with changing F_0 . (ii) The quantum efficiency of luminescence can be a function of the excitation laser power $\eta = \eta(F)$, which also influences on the properties of integral luminescence according to (2).

As far as the expression (5) is derived from the assumption $\eta = \text{const}$, it accounts for the non-linearity of integral luminescence caused by the non-linearity of absorption. This fact provides a principal possibility to separate the effect of non-linear absorption from that of a non-constant quantum efficiency. If an experiment reveals the violation of expression (5), it can be considered as an indication of laser-induced changes in the luminescence quantum efficiency. Below we consider a couple of examples illustrating the use of the expression (5).

2.1. Saturated absorption

Consider a simple two-level model of molecular luminescence by means of the following balance equation

$$\frac{dn_2}{dt} = \sigma F n_1 - \frac{n_2}{\tau}, \quad (7)$$

where σ is the absorption cross-section, τ is the luminescence lifetime, n_1 and n_2 are the occupancies of the ground and excited states, respectively, $n_1 + n_2 = N$ is the numerical density of molecules. Using a stationary approximation, the luminescence power emitted from an elementary volume dV can be written as

$$dP_L = \hbar\omega \frac{n_2}{\tau} dV = \hbar\omega \frac{N}{\tau} \cdot \frac{F}{F + F_{\text{sat}}} S dz, \quad (8)$$

where $F_{\text{sat}} = (\sigma\tau)^{-1}$. Besides, the difference of the laser power surface density dF passing through the layer of the thickness dz can be written [5] as a non-linear photon transport equation

$$dF = -\sigma F n_1 dz = -\sigma F N \frac{F_{\text{sat}}}{F_{\text{sat}} + F} dz. \quad (9)$$

By substituting (9) into (8) and integrating the luminescence power over the whole volume, $V = Sd$, we obtain the integral power of luminescence as follows

$$P_L = \int_{F_0}^{F_d} \hbar\omega S \frac{dF}{\sigma\tau F_{\text{sat}}} = \frac{\hbar\omega S F_0}{\sigma\tau F_{\text{sat}}} (1 - T). \quad (10)$$

By differentiating the expression (10), we can make sure of that the expression (5) holds true.

The solution of the transport equation (9) can be given in the following implicit form

$$\frac{F_d}{F_0} \exp\left(\frac{F_d}{F_{\text{sat}}}\right) = T_0 \exp\left(\frac{F_0}{F_{\text{sat}}}\right), \quad (11)$$

where T_0 is the low-signal transmittance, i.e., the transmittance without saturation. The parameter of the transmission non-linearity γ_T can be easily derived from (11) as follows

$$\gamma_T = \frac{1 + F_0/F_{\text{sat}}}{1 + F_d/F_{\text{sat}}} - 1. \quad (12)$$

The expressions (12) and (5) yield the following expression for γ -parameter of integral luminescence

$$\gamma_L = \frac{1}{1 + F_d/F_{\text{sat}}}. \quad (13)$$

As seen from (13), γ_L depends on F_0 due to the dependence $F_d(F_0)$ that can be calculated by numerical solution of the equation (11). The results of calculations performed for the expressions (11)–(13) are given in Fig. 1a, curves 1-3.

It is worth noting that the above expressions are valid under the condition of uniform distribution of the laser power density across the beam and in the stationary approximation. As mentioned in [4], if the investigated process is characterized with a power-dependent parameter of non-linearity, significant errors arise due to the non-uniformity of spatial distribution of the laser power. Thereinafter, by means of computer simulation, we consider the effect of the mentioned error-causing factors on the validity of the expression (5).

For modelling the non-uniform distribution of the power density across the laser beam, we use the following Gauss function

$$F(r) = F_0 \exp(-r^2\Delta^{-2}),$$

where γ is the distance from the beam axis, Δ is the beam radius. Then the expression (3) is transformed into the following

$$P_L = \hbar\omega\eta \int_0^\infty [F_0 \exp(-r^2\Delta^{-2}) - F_d(r)] 2\pi r dr. \quad (14)$$

The distribution of transmitted laser power $F_d(\gamma)$ is an unknown function to be calculated.

The parameter γ_L of integral luminescence was calculated by numerical integration of the expression (14). While integrating, the unknown function $F_d(\gamma)$ was determined by numerical solution of the equation (11) by substitution of $F_0 \exp(-r^2\Delta^{-2})$ for F_0 . The calculated dependence $\gamma_L(F)$ is shown in Fig. 1a, curve 6. Fig. 1a also presents the calculated curves $\gamma_F = 1 + \gamma_T$ and T (curves 5 and 4, respectively). While calculating γ_T and T , the transmitted laser power $F_d(\gamma)$ was integrated over the beam cross-section using (11).

By substitution of the calculated γ_T and T into (5), we obtain the dependence $\gamma_L(F_0)$ that can be compared with that calculated using (14). The results of calculations show that both mentioned curves $\gamma_L(F_0)$ coincide within the accuracy of approximation $\Delta\gamma_L \leq 0.0001$. The coinciding curves $\gamma_L(F_0)$ are plotted in Fig. 1a as the single curve 6. Thus, the calculations confirm the validity of the expression (5) in the case of non-uniform distribution of the laser power density across the beam.

Now we consider the expression (5) in the case of non-stationary excitation of molecular luminescence. Taking into account that photodetectors often operate in the integrating mode, i.e., measuring an integral of optical pulse over time, we calculate the energy of luminescence and laser pulse as follows

$$\varepsilon_L = \frac{\hbar\omega}{\tau} \int_{-\infty}^\infty dt \int_V dV n_2, \quad (15)$$

$$\varepsilon_F = \int_{-\infty}^\infty dt \int_S dS F.$$

While integrating, the integrands n_2 and F were calculated numerically from the equations (7) and (11). The temporal shape of the laser pulse was given by the following function

$$F(r, t) = F(r) \exp(-t^2\tau_i^{-2} 4 \ln 2), \quad (16)$$

where τ_i is the laser pulse duration. Calculations were performed for two examples of luminescence lifetime, $\tau = 6$ ns and 30 ns, and for the laser pulse duration $\tau_i = 10$ ns. The results of calculation are shown in Fig. 1b. Again we calculated $\gamma_L(F_0)$ by two ways from the expressions (5) and (15) and both the curves prove to coincide. In Fig. 1b, these coinciding curves are plotted as a single one (curve 9 for $\tau = 6$ ns and curve 12 for $\tau = 30$ ns). Thus, we demonstrate the validity of the expression (5) in the non-stationary case.

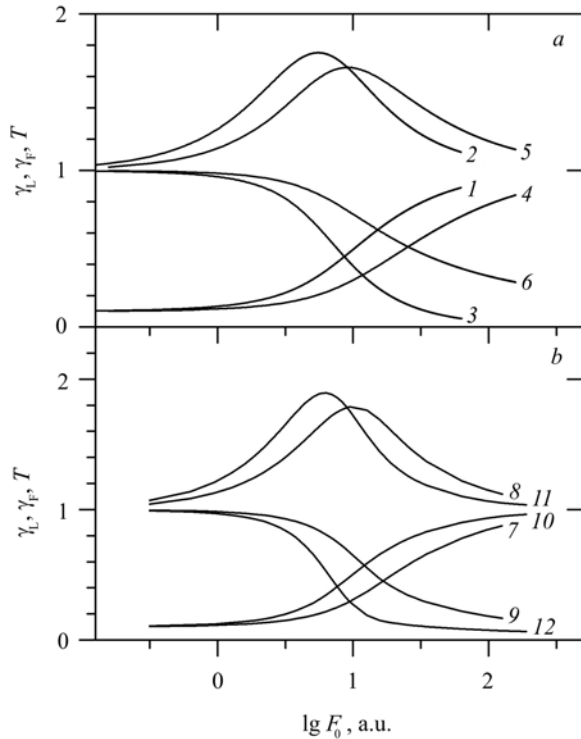


Fig. 1. The results of calculation of optical transmittance T (1, 4, 7, 10), parameters of non-linearity $\gamma_F = \gamma_T + 1$ (2, 5, 8, 11) and γ_L (3, 6, 9, 12) as functions of F_0 for uniform (1–3) and Gauss (4–6) distribution of the laser power density across the beam, for the stationary (1–6) and non-stationary (7–12) approximations with $\tau = 6$ ns (7–9) and 30 (10–12).

As seen from Fig. 1, the non-uniformity of laser power distribution and the non-stationary behavior of occupancies cause significant changes in the curves $\gamma_L(F_0)$, $\gamma_F(F_0)$, and $T(F_0)$ (for example, the γ_L -values calculated for different models are several times different for the same F_0). Nevertheless, the expression (5) remains valid.

For experimental verification of the expression (5), we measured the laser-induced luminescence and transmittance of an aqueous solution of Rhodamine 6G. As an excitation source, the second harmonic of a Q-switched YAG:Nd³⁺ laser was used (wavelength $\lambda = 532$ nm, $\tau_i = 10$ ns). The luminescence was detected at the fixed wavelength 585 nm (through a single grating monochromator). The low-signal transmittance was $T_0 = 0.1$. The luminescent track was completely located within the operating field of the photodetector. Measurements were performed at room temperature. The results of measurements are plotted in Fig. 2 where open circles represent γ_L calculated in accord to the definition (4) by using luminescence power experimental data, and filled circles are result of calculations according to the expression (5) with the experimental data taken for T and γ_L . As seen from the figure, the curves 3 and 4 are in good agreement, which substantiates the validity of the expression (5).

2.2. Two-photon absorption

Now we consider the mechanism of two-photon absorption of laser radiation by molecules or luminescence centers. This mechanism implies transitions through the intermediate virtual states of molecules with simultaneous absorption of a couple of photons. When a powerful laser radiation interacts with molecules that absorb laser light by the two-photon mechanism the decrease of the propagating laser beam power can be described as follows [5]

$$dF = -\beta F^2 dz$$

with the well-known solution

$$F(z) = F_0 (1 + \beta F_0 z)^{-1}, \quad (17)$$

where β defines the probability of two-photon transitions in the given centers/molecules. From the expression (17), the parameter of non-linearity can be easily derived

$$\gamma_F = (1 + \beta F_0 d)^{-1}. \quad (18)$$

Suppose the two-photon absorption excites luminescence of molecules. For an elementary volume dV , the power of luminescence emitted in all directions is proportional to the square of laser power density, $dP_L = \text{const } F_2 dV$. Then the integral power of luminescence is

$$P_L = \text{const } F_0^2 (1 + \beta F_0 d)^{-1} S d.$$

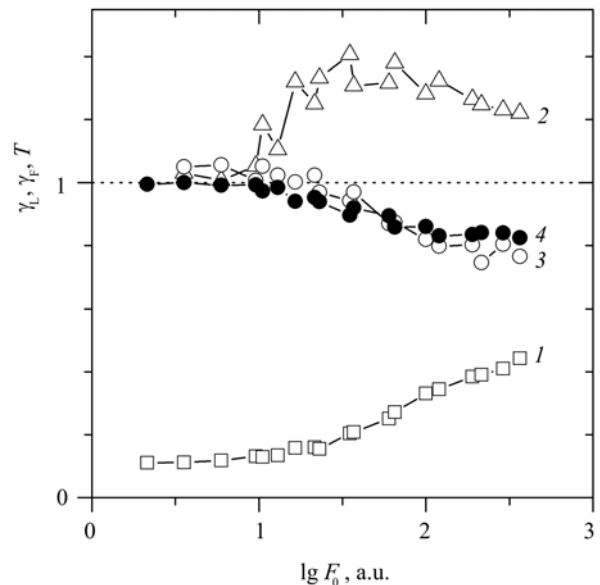


Fig. 2. Optical transmittance T (1) and parameters of non-linearity γ_F (2) and γ_L (3, 4) of aqueous solution of Rhodamine 6G as a function of the excitation laser power. Filled circles are the values of γ_L calculated with (5) using the experimental data $T(F_0)$ and $\gamma_T(F_0)$.

Comparing this expression with (17), we see that $P_L \sim F_0 F_d$. This fact yields the following simple relation for the parameters of non-linearity

$$\gamma_L = 1 + \gamma_F = 2 + \gamma_T. \quad (19)$$

The expression (19) seems to be unusual as it predicts that γ -parameter of integral luminescence excited via two-photon absorption can be different from its well-known value of 2. This difference can take place when $\gamma_F \neq 1$ or $\gamma_T \neq 0$, i.e., when the two-photon absorption essentially changes the distribution of the laser power within the luminescent volume. This is an example of the effect of non-linear two-photon absorption on the characteristics of luminescence. However, this case is difficult to observe experimentally, as the probability of two-photon absorption is usually low (in most of molecules and impurity centers).

Now we consider the situation when a luminescence center absorbs two laser photons in sequence (step by step) through an intermediate stationary state. Such a sequential absorption is usually much more probable than two-photon absorption through a virtual state. Sequential absorption can result in the excitation of luminescence together with the significant non-linear depletion of the laser beam.

Consider the following model of sequential absorption in an impurity center (Fig. 3) where three energy levels (1, 2, and 3) are involved. In Fig. 3, σ_{12} and σ_{23} are the absorption cross-sections, τ_2 and τ_3 are the luminescence lifetimes, Q represents the non-radiative relaxation. Suppose the probability of relaxation exceeds the rate of laser excitation, $Q \sim \sigma_{12}F, \sigma_{23}F$. Denote the occupancies of levels 1, 2, and 3 as n_1, n_2, n_3 , respectively. Suppose the laser-induced decrease of occupancy of ground state is negligible, i.e., $n_2, n_3 \sim n_1 \approx N$. Besides, suppose the occupancy of the

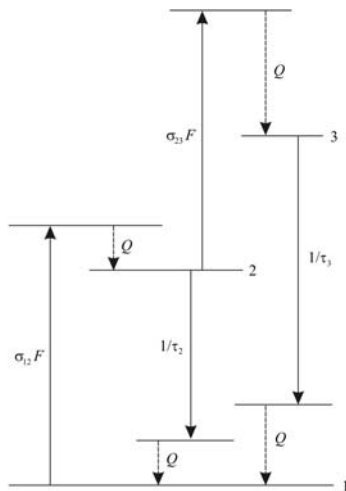


Fig. 3. Optical transitions in a luminescence center.

first excited state is proportional to the laser power density, $n_2 \sim F$. (Though this supposition seems to be intuitively obvious, it requires some argumentation, which will be given thereafter.)

According to the above model, the decrease of power of the propagating laser beam can be described

$$dF = -(\alpha F + \beta F^2) dz, \quad (20)$$

where $\alpha = \sigma_{12}N$ is the absorption coefficient, β is a constant proportional to the cross-section σ_{23} . By integrating (20) we obtain

$$F(z) = F_0 \left[1 + \frac{\beta}{\alpha} F_0 (1 - \exp(-\alpha z)) \right]^{-1} \exp(-\alpha z), \quad (21)$$

$$\gamma_F = \left[1 + \frac{\beta}{\alpha} F_0 [1 - \exp(-\alpha d)] \right]^{-1}. \quad (22)$$

From (21) when $z = d$ and (22), it follows

$$T T_0^{-1} = \gamma_F. \quad (23)$$

According to the model (Fig. 3), the power of luminescence from the level 3 emitted from a unit volume is proportional to the square of the excitation laser power. Then for the integral power of luminescence from the level 3, taking into account the expression (21), the integration over the volume leads to the following relation $P_L \sim F_0 F_d$, hence it follows that the parameters of non-linearity obey the expression (19), similarly to the case of two-photon excitation.

Now we consider some reasoning in respect to the relation $n_2 \sim F$. First, in the case of stationary approximation ($\tau_2 \sim \tau_i$), the following balance equation

$$\frac{dn_2}{dt} = 0 = \sigma_{12}FN - \frac{n_2}{\tau_2} - \sigma_{23}Fn_2$$

shows that the relation $n_2 \sim F$ requires proportionality between n_2 and F under the condition that the third term in the right-hand side of the equation is negligible as compared with the second term. This implies $F < (\sigma_{23}\tau_2)^{-1}$. This condition can be easily fulfilled experimentally by limiting the laser power; however, it will limit the range of observable changes of optical transmission at a level of several percents. With such limitations, the calculations of γ -parameters using the experimental data become difficult because of the fluctuations of the laser power. Second, if the stationary approximation break down ($\tau_2 \geq \tau_i$), then the above-mentioned condition of limitation of the laser power is written as follows $F < (\sigma_{23}\tau_i)^{-1}$ with the same consequences. Moreover, without the stationary approximation, calculations of optical signals require integration over time, which makes it practically impossible to obtain a simple analytical relation for the parameters of non-linearity. Thus, we note that the above expressions for the parameters γ_T and γ_L , obtained from

the model of sequential absorption of two laser photons should be considered as a first approximation.

For experimental verification of the main relations used in this section, the luminescence of YAG:Nd³⁺ crystals was investigated. As known, the absorption and luminescence spectra of YAG:Nd³⁺ crystals contain a large number of narrow lines in the wide spectral range [6]. Most of these lines are assigned to the transitions between the energy levels (split in the crystal field) of impurity ions Nd³⁺ with the electron configuration 4f³. In Fig. 3, the lower level 1 represents ⁴I_{9/2} level of Nd³⁺ ion. In this work, the third harmonic of YAG:Nd³⁺ laser (wavelength $\lambda = 355$ nm) was used to excite luminescence of YAG:Nd³⁺ crystal. For this excitation wavelength, the level 2 in Fig. 3 corresponds to the level ²P_{3/2}, and the level 3 – to the level ²F_{5/2} of Nd³⁺ ion [7]. High excited states, from which the non-radiative relaxation occupies the level ²F_{5/2}, are assigned to 4f²5d configuration of Nd³⁺ ion [8]. *f* → *fd* transitions in Nd³⁺ ions are relatively strong, hence the appropriate cross-section σ_{23} is enough high to make a chance for easy experimental observation of significant non-linear absorption. The rate of non-radiative relaxation (Q in Fig. 3) can be approximately estimated as 10⁹ s⁻¹ [9]. The luminescence from ²F_{5/2} level includes a number of lines in the visible spectral range, all of them are characterized with the lifetime $\tau_3 \approx 3$ μ s. In this paper, the luminescence from ²F_{5/2} level was detected at the wavelength close to 401 nm that corresponds to ²F_{5/2} → ²H_{9/2} transition in Nd³⁺ ion.

The results of experiments are shown in Figs 4 and 5. As seen from the figures, the increase of laser power causes significant decrease of the parameters γ_L at $\lambda = 401$ nm (curve 1 in Fig. 4) and γ_F (curve 1 in Fig. 5). Such behavior is in agreement with the above-

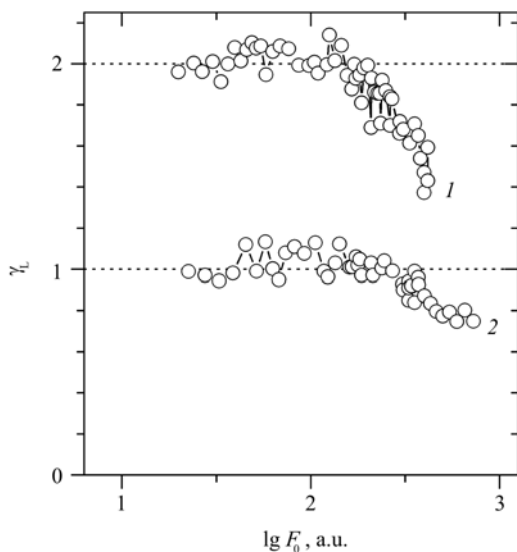


Fig. 4. Parameters γ_L of integral luminescence of YAG:Nd³⁺ crystal, measured at the wavelengths 401 nm (1) and 740 (2), as functions of the excitation laser power.

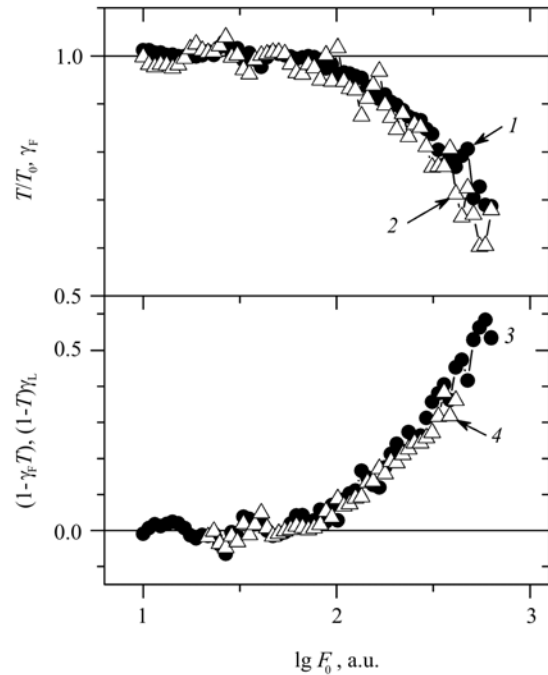


Fig. 5. Parameter γ_F (1, filled circles), normalized transmittance T/T_0 (2, triangles), $(1 - T)\gamma_F$ (3, filled circles), and $(1 - T)\gamma_L$ at $\lambda = 401$ nm (4, triangles) of YAG:Nd³⁺ crystal as functions of the excitation laser power.

considered model. Besides, the agreement between the parameter γ_F and the normalized transmittance T/T_0 (Fig. 5, curves 1 and 2, respectively) supports a validity of the expression (23).

To verify the expression (5), we used Fig. 5 presenting the plots of appropriate combinations of experimental data $(1 - T)\gamma_F$ (curve 3) and $(1 - T)\gamma_L$ at $\lambda = 401$ nm (curve 4). The agreement of curves 3 and 4 confirms the validity of the expression (5).

As seen from Figs 4 and 5, the observed decrease of γ_L at $\lambda = 401$ nm with F_0 is larger than the appropriate decrease of γ_F . This fact contradicts to the expression (19). According to (19), it is expected that the curve $\gamma_F(F_0)$ should coincide with the curve $\gamma_L(F_0)$ being shifted along the ordinate by a unity. The observed disagreement between the theory and experiment can be explained if we suppose that the conditions of experiments do not fulfil the above-mentioned relation $F < (\sigma_{23}\tau_i)^{-1}$. The results of the following experiment confirm the validity of this supposition. We investigated luminescence from the level 2 (Fig. 3) that corresponds to ²P_{3/2} level of Nd³⁺ ion. This luminescence was detected at the wavelength 740 nm that corresponds to ²P_{3/2} → ⁴F_{5/2} transitions. As seen from Fig. 4 (curve 2), γ_L at $\lambda = 740$ nm decreases with F_0 , which indicates violation of the relation $n_2 \sim F$ at high levels of laser excitation. Thus, we conclude that the observed decrease of γ_L at $\lambda = 401$ nm with F_0 (Fig. 4, curve 1) is caused

by at least two reasons: (i) sequential absorption without the saturation of the transition $1 \rightarrow 2$ (according to (19)), and (ii) saturation of the transition $1 \rightarrow 2$ (violation of the relation $n_2 \sim F$).

Thus, the results presented in Figs 4 and 5 confirm the validity of theoretical relations between the parameters of non-linearity, at least to a first approximation.

3. Concluding remarks

This paper presents the analysis of the effect of non-linear absorption on the characteristics of laser-induced luminescence. For characterization of degree of non-linearity, both for luminescence and transmittance, we propose to use similar dimensionless parameters, γ_L (4) and γ_T (6) or γ_F , which can be easily calculated from the experimental data. For the case of the constant quantum efficiency of luminescence, the universal expression (5) is derived, which gives a useful relation for the mentioned parameters of non-linearity of absorption and luminescence. The validity of the expression (5) is verified for two examples of non-linear absorption and luminescence: saturated molecular luminescence and luminescence of impurity centers excited via sequential absorption of two laser photons.

References

1. E.N. Arkad'eva, O.A. Matveev, S.M. Ryvkin, Yu.V. Rud' // *Sov. Phys.-Tech. Phys.* **36**, p. 1146 (1966).
2. E.N. Arkad'eva, O.A. Matveev, S.M. Ryvkin, Yu.V. Rud' // *Sov. Phys. Semicond.* **1**, p. 805 (1967).
3. A. Khusainov, R. Arit, P. Siffert // *Nucl. Instr. and Meth.* **A380**, p.245 (1996).
4. M.R. Squillante, G. Entine, E. Frederick, L. Cirignano, T. Hazlett // *Ibid.* **A283**, p. 323 (1989).
5. T. Takahashi, K. Hirose, C. Matsumoto *et al.* // *Proc. SPIE*, **3446**, p. 29 (1998).
6. C. Matsumoto, T. Takahashi, K. Takizawa *et al.* // *IEEE Trans. Nucl. Sci.* **45**, p. 428 (1998).
7. T. Takahashi, B. Paul, K.Hirose *et al.* // *Nucl. Instr. and Meth.* **A436**, p. 111 (2000).
8. T. Takahashi, S. Watanabe, G. Sato *et al.* // *IEEE Trans. Nucl. Sci.* **48**, p. 287 (2001).
9. T. Takahashi, S. Watanabe // *Ibid.* **48**, p. 950 (2001).
10. Home page of Amptek Inc. www.amptek.com
11. S. Sze, *Physics of semiconductor devices*, 2nd ed., Wiley, New York (1981) p. 245.
12. D.M. Hofmann, W. Stadler, P. Chrismann, B.K. Meyer, Defects in CdTe and Cd_{1-x}Zn_xTe // *Nucl. Instr. and Meth.* **A380**, p. 117 (1996).
13. R. Grill, J. Franc, P. Höschl *et al.* // *IEEE Trans. Nucl. Sc.* **49**, N3, p. 1 (2002).
14. L.A. Kosyachenko, A.V. Markov, S.E. Ostapov *et al.* // *Semiconductors* **36**, p. 1218 (2002).
15. L.A. Kosyachenko, A.V. Markov, Ye.L. Maslyanchuk *et al.* // *Ibid.* **37**, p. 1373 (2003).