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## Some Aspects of the Inverse Problem of Determination of $\text{Cr}^{4+}$ :YAG Absorption Cross Sections Using Experimental Data of Transmission

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### Abstract

Mathematical modelling of nonlinear transmission of  $\text{Cr}^{4+}$ :YAG crystals excited by Nd:YAG laser pulses of different duration are reported. Numerical simulation of transmission using a five-level scheme which included finite excitation lifetimes in  $\text{Cr}^{4+}$ :YAG, focusing and diffraction of transversely nonhomogeneous pump radiation allowed to specify more precisely the limits of applicability of the four-level model. Critical analysis of typical simplifications commonly used for solving an inverse problem of determination of absorption cross sections is undertaken. It is shown that some of these assumptions that seem plausible enough might lead to considerable deviations of the determined absorption cross sections from their "true" values. A technique for determination of confidence intervals in case of the nonlinear regression is emphasized.

**Keywords:** transmission saturation, inverse problem, absorption cross sections

### 1 Introduction

Solution of an inverse problem consists of determination of the parameters of the utilized mathematical model from the available experimental data [1,2]. The inverse problems are very widely used in physics. They need to be solved in order to obtain various kinds of information, e.g., solution of an inverse problem of bleaching of the dyes is used for measurement of duration of the

picosecond pulses [3], the results of gain saturation are used for determination of the lifetimes of active laser media [4] and optimization of the input pulse parameters in order to obtain the desired temporal shape of the amplified pulse [5], etc. However, as the methods proposed by the mathematicians for processing the experimental data and solving the inverse problems [6,7] seem to be too complex for most of the physicists, the methods usually employed by them for processing the experimental data contain many shortcomings resulting in significant errors of determination of the parameters being searched for. Besides, the issue of the confidence intervals of the determined values of the parameters usually remains unresolved. Clarification of this issue is currently receiving much attention. For instance, to conform the international guide to the expression of uncertainty, the uncertainty of a measurement in a wide sense means a doubtfulness concerning the confidence of the measurement result [8].

In this paper, we shall analyze the situation in processing the experimental data using as an example the problem of determination of the ground and excited absorption cross sections of the  $\text{Cr}^{4+}$ :YAG crystal from the experimental data of transmission in the absorption saturation regime (see [9-15] and references therein), and demonstrate that the seemingly plausible enough “simplifications” often lead to considerable deviations of the obtained cross sections from the “true” ones, i.e., in fact to an erroneous solutions of the inverse problem. A technique for determination of confidence intervals in case of the nonlinear regression will be emphasized. We shall also note that the situation with determination of the parameters of nonlinear media from the experimental saturation curves is quite typical and can be often found in other physical problems, like determination of cross sections of the stimulated emission of the laser media [16,17], saturation fluorimetry [18], etc.

## 2 Modelling of $\text{Cr}^{4+}$ :YAG Transmission Using a Five-Level Scheme

In recent years there has been a considerable interest in using  $\text{Cr}^{4+}$ :YAG for passive  $Q$ -switching of the Nd-doped lasers. For the purpose of mathematical modelling of generation dynamics of this type of lasers, it is necessary to know the values of absorption cross sections and lifetimes of the excited levels with sufficient precision [19-21]. Lasers with a short cavity and especially the microchip lasers permit to obtain pulses of nano- and subnanosecond duration. At the same time, investigations of bleaching and measurements of the  $\text{Cr}^{4+}$ :YAG cross sections have usually been carried out using the lasers with long ( $\tau_l \geq 20$  ns) pulses [10,14,15]. Therefore, for modeling of bleaching in these cases, it is assumed that the decay times of the upper excited levels are very fast and a simplified four-level scheme is used even in cases when the trains of picosecond pulses are used for exciting [12].

A recent paper [11] proposed to use the five-level scheme for description of energy levels of  $\text{Cr}^{4+}$ :YAG that is widely used for modeling of saturable absorption and reverse saturable absorption of complex molecular compounds [22]. However, the experimental investigation of nonlinear absorption of  $\text{Cr}^{4+}$ :YAG has been carried out using the Nd:YAG lasers with greatly differing pulsewidths of  $\tau_{L1} \sim 40$  ps and  $\tau_{L2} \sim 25$  ns [11]. Therefore, as acknowledged by the authors themselves, the simulation was not very sensitive to the small change in the relaxation time  $\tau_3$  of the excited state  ${}^3T_{1g}$  and the intersystem crossing time  $\tau_{24}$ . It is hard to estimate the extent of reliability of above mentioned models without additional theoretical investigations. Therefore, the experimental investigations and corresponding simulations of transmission of  $\text{Cr}^{4+}$ :YAG in the range of subnano- and nanosecond pulses are of great importance.

We shall also note that all the previous papers modelled the transmission of  $\text{Cr}^{4+}$ :YAG in the plane-wave approximation. On the other hand, the experimental investigations usually involve rather tight beam focusing for obtaining high energy densities. For instance, for the experimental setup described in Ref. [11], the beam diameter at the sample is only  $2w_0 = 35$   $\mu\text{m}$ . Therefore, the length of the beam waist  $L_D = k_0 w_0^2 / 2 \approx 3.3$  mm is comparable to the sample thickness  $l = 1.94$  mm (here,  $k_0 = 2\pi n / \lambda$  is the wavenumber,  $n = 1.82$  is the refraction index and  $\lambda = 1064$  nm is the wavelength of the Nd:YAG laser). Additional small diaphragms sometimes utilized in order to obtain rectangular intensity distribution lead to significant diffraction distortions of the field structure. All these factors contribute to significant difficulties in processing the experimental data and they are one of the reasons why the published data about the ground and excited state absorption cross sections differ largely. The absorption cross sections might generally depend on the technology of the sample production, but deviations of the values of absorption cross sections brought about by the technological differences are usually not so critical [12-14]. Therefore, in our opinion, the reasons of large deviations of the published data of the  $\text{Cr}^{4+}$ :YAG absorption cross sections are differences in processing the experimental data.

Numerical simulation of transmission of the five-level saturable absorber (SA) when using tightly focused laser beams must be done using the equation for the slowly-varying complex amplitude of the laser pulse, including the term describing focusing and diffraction of the beam [13]. The following set of equations expressed in the dimensionless variables describes the pulse propagation in the SA:

$$\frac{\partial e}{\partial t} + \frac{\partial e}{\partial z} - iD\Delta_r e + \frac{\alpha}{2}e = -\frac{1}{2}(\sigma_o n_1 + \sigma_{e1} n_2 + \sigma_{e2} n_4)e, \quad (1)$$

$$\frac{\partial n_1}{\partial t} = -\sigma_o n_1 |e|^2 + \frac{n_2}{\tau_2} + \frac{n_4}{\tau_4}, \quad (2)$$

$$\frac{\partial n_2}{\partial t} = \sigma_o n_1 |e|^2 - \sigma_{e1} n_2 |e|^2 + \frac{n_3}{\tau_3} - \frac{n_2}{\tau_2} - \frac{n_2}{\tau_{24}}, \quad (3)$$

$$\frac{\partial n_3}{\partial t} = \sigma_{e1} n_2 |e|^2 - \frac{n_3}{\tau_3}, \quad (4)$$

$$\frac{\partial n_4}{\partial t} = -\sigma_{e2} n_4 |e|^2 + \frac{n_2}{\tau_{24}} + \frac{n_5}{\tau_5} - \frac{n_4}{\tau_4}, \quad (5)$$

$$\frac{\partial n_5}{\partial t} = \sigma_{e2} n_4 |e|^2 - \frac{n_5}{\tau_5}, \quad (6)$$

$$n_1 + n_2 + n_3 + n_4 + n_5 = n_0. \quad (7)$$

Here, the longitudinal and transverse coordinates are normalized to the sample length  $l$  and the radius  $r_0$ , the time is normalized to the sample transit time  $t_0 = l/v$  ( $v$  is the speed of the light in the sample). The field amplitude  $e$  and the population densities  $h_i$  are normalized to  $E_0$  (corresponding to  $I_0 = hv/\sigma t_0$ ) and  $\bar{N} = (\sigma l)^{-1}$  respectively,  $\sigma = 8.8 \times 10^{-19} \text{ cm}^2$  is the effective spectroscopic emission cross section of the Nd:YAG. Besides, the following notations are introduced:  $\Delta_r = \partial^2 / \partial r^2 + \partial / r \partial r$ ,  $D = l/2k_0 r_0^2$ ,  $\alpha = \alpha_0 l$  is the dimensionless nonresonant absorption coefficient. For brevity, the same notations are preserved for the normalized absorption cross sections from the ground  $\bar{\sigma}_0 = \sigma_0 / \sigma$  and excited  $\bar{\sigma}_{e1,2} = \sigma_{e1,2} / \sigma$  levels, as well as those for the dimensional and dimensionless relaxation times, beam radii, pulse energies, etc.

It is well known that the transmission of Cr<sup>4+</sup>:YAG depends on the mutual orientation of the polarization of radiation and the crystallographic axes [9-15]. However, this dependence is often neglected. Equations (1)–(7) correspond to the specific case of the laser pulse propagation along the crystallographic axis [001] with linear polarization aligned along the crystallographic axis [100] or

[010]. Therefore, in order to correctly compare the theoretical results with the experiments, the SA sample must be aligned accordingly. Note also that equations (1) – (7) do not include the possibility of absorption by the  $\text{Cr}^{4+}$  centers aligned across the radiation polarization. This so-called cross-saturation in the absorber is rather small [23]. The values of the corresponding absorption cross sections do not exceed 5% of the values of absorption with the parallel alignment and fall within the errors of measurement of the latter [14].

Equations (1) – (7) were solved using the Crank-Nicholson finite-difference method with splitting by the physical factors [24] with the following initial and boundary conditions:

$$e(r, z, t = 0) = 0, \quad n_1(r, z, t = 0) = n_0, \quad n_{2-5}(r, z, t = 0) = 0, \quad (8)$$

$$e(r, z = 0, t) = e_0 f(t) \exp[-(r/w_0)^S - i\varphi(r)]. \quad (9)$$

The calculations have been carried out for the factorized pulses (9) with the Gaussian or super-Gaussian ( $S = 2, 4, 6, \dots$ ) transverse intensity distribution taking into account the focusing or the beam aberration by means of the function  $\varphi(r)$ . The short pulses with steeper leading edges obtained by using the SBS-compression [13] were described by the function

$$f(t) = [(2e)^{1/2} t / \mu\tau_L] \exp[-t^2 / (\mu\tau_L)^2], \quad \mu = 1.2245. \quad (10)$$

The calculations were performed with different beam profiles and various pulse shapes using the experimental values of the sample thickness  $l = 4.2$  mm and the Gaussian beam radius  $w_0 = 0.24$  mm. More intense absorption on the leading edge of the pulse [11] results in general steepening and shortening of the leading edge of the pulse after traversing the SA. Shortening of the output pulse at the half maximum of the instantaneous power is not monotonous as the incident pulse energy grows. The minimum duration measured by the intensity on the beam axis and by the total instantaneous power of the beam is obtained at different values of the incident pulse energies.

### 3 Solving the Inverse Problem Using the Four-Level Model

Experimental measurements of the transmission have been carried out utilizing the Nd:YAG laser system with an SBS-compressor [13]. By varying the focusing conditions, the duration of the output pulse was tuned in the range from 0.15 to 2.5 ns. The results of transmission measurements are shown in

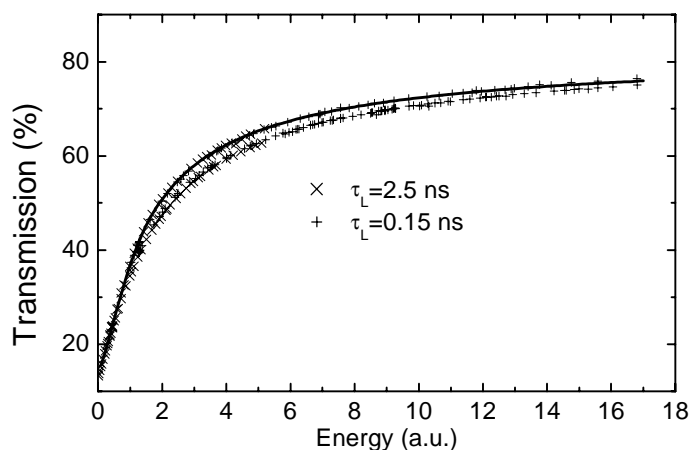


Fig. 1. Energy transmission of the  $\text{Cr}^{4+}$ :YAG crystal for nanosecond ( $\times$ ) and subnanosecond (+) durations of pump pulses for parallel ( $\theta = 0^\circ$ , upper set of points) and tilted ( $\theta = 45^\circ$ , lower set of points) crystallographic axes with respect to linear polarization of the incident radiation. The solid line denotes the theoretical transmission curve for optimum cross sections.

Fig. 1. In case of the polarization direction coinciding with that of the crystallographic axis (the tilt angle  $\theta = 0^\circ$ ), the maximum transmission was observed (the upper set of the experimental points). In case when the SA was rotated by the angle  $\theta = 45^\circ$ , the minimum transmission (the lower set) was observed. No dependence of the transmission on the pulse duration was observed. In both series of measurements, the values of transmission denoted by different symbols for pulse duration of  $\tau_L = 2.5 \text{ ns}$  and  $\tau_L = 150 \text{ ps}$  are undistinguishable. As it can be seen in Fig. 1, the difference in the transmission curves for different orientations of the SA seems to be insignificant at a first glance. However, the calculations of the generation dynamics [20,21] show that in some cases inclusion of the transmission anisotropy might result in changes in the duration of the generated pulses by an order of magnitude when the SA is rotated with respect to the crystallographic axis [001] coinciding with the optical axis of the generator.

The theoretical energy transmission of the SA is determined by

$$T = \int_0^{\infty} u(r,1) r dr / \int_0^{\infty} u(r,0) r dr, \quad (11)$$

where  $u(r, z) = \int_{-\infty}^{\infty} |e(r, z, t)|^2 dt$  is the energy density of the pulse normalized to

$h\nu/\sigma$ . Solution of the direct problem, i.e., calculation of the transmission depending on the incident energy at the given parameters of the media and the pulse, is quite straightforward. Certain assumptions concerning the excitation relaxation times are necessary for calculation of the transmission. In the papers that are known to us (see [9-15] and references therein), the fast relaxation from the excited levels is mostly assumed, or, based on certain assumptions that are not completely substantiated, it is stated that either  $\tau_{24} = 0$  and  $\tau_3 = 0.1$  ns [12],  $\tau_3 = 0.55$  ns [10], or  $\tau_{24} \sim 4$  ns and  $\tau_3 = 10$  ps [11]. Using these values of the relaxation times, we could not achieve an agreement between the calculated values of transmission with the experimental ones (the latter did not depend on the duration of the exciting pulse ( $\tau_L = 150$  ps or  $\tau_L = 2.5$  ns) by selecting the values of the cross-sections. The durations of the laser pulses used in the experiments were close enough to the specified relaxation times. Therefore, for the cases where the calculated and experimental transmission values were in agreement for the short exciting pulses, the calculated transmission values were significantly different from the experimental ones in case of the longer exciting pulses, and vice-versa.

On the other hand, solution of the inverse problem – determination of the parameters of the SA from the experimental transmission data – requires calculation of the transmission (11) for a large number of the sets of the SA parameters at various incident pulse energy values corresponding to the experimental data. Besides, solution of the inverse problem by minimizing the function that evaluates the mean square deviations of the calculated values from the measured ones might have several local minima in case of the presence of many variables [18]. This factor significantly aggravates the minimization process. In this situation, the time of solution of the direct problem with sufficient accuracy becomes a critical factor requiring to simplify the utilized model. For that case, neglecting the diffraction ( $D \rightarrow 0$ ) when using sufficiently broad beams ( $L_D \gg l$ ) and also assuming  $\sigma_{e1} \equiv \sigma_e$ ,  $\tau_3 \rightarrow 0$  and slow cross-relaxation ( $\tau_L \ll \tau_{24}$ ) to the neighboring set of levels, it is easy to obtain from equations (1) – (7) the standard four-level scheme of the SA. It can be also seen that the equivalent and commonly used four-level scheme can be obtained from the five-level one by setting also  $\sigma_{e1} = 0$ ,  $\sigma_{e2} \equiv \sigma_e$  and

$\tau_{24}, \tau_5 \rightarrow 0$ . Using these approximations, and following the technique described in [9], the following equation governing the variation of the pulse energy density inside the SA can be obtained:

$$\frac{du(r, z)}{dt} = \frac{\gamma_1 - \gamma_2}{\sigma_0} [\exp(-\sigma_0 u(r, z)) - 1] - \gamma_2 u(r, z). \quad (12)$$

The transmission in every point  $(r, z)$  of the beam is determined by only three independent parameters of the SA:  $\sigma_0$ ,  $\gamma_1 = n_0 \sigma_0 + \alpha$ , and  $\gamma_2 = n_0 \sigma_e + \alpha$ . We shall note that a recent paper [14] also includes additively in an equation of a similar type the terms describing the absorption from the phototropic centers aligned perpendicularly to the linear polarization vector of the exciting beam. Therefore, it is not quite clear how the contributions of the different terms can be discriminated when solving the inverse problem of determination of the absorption cross-section. In our opinion, introduction of similar terms is not completely substantiated, especially as the values of the absorption cross sections obtained in [14] for the radiation with perpendicular polarization is by an order of magnitude smaller than the values of absorption cross sections for the radiation with parallel polarization and fall within the error limits of the latter.

For sufficiently broad beams with the rectangular transverse distribution of the energy density  $u_0$ , the following relations for the transmission can be obtained [9]:

$$1/T \cong a + bu_0, a = \exp(\gamma_1), b = -\frac{1}{2} \sigma_0 (1 - \gamma_2 / \gamma_1)(a - 1), \quad u_0 \ll 1, \quad (13)$$

$$T \cong c + d/u_0, c = \exp(-\gamma_2), d = -(\gamma_2 / \gamma_1 - 1)(1 - c) / \sigma_0, \quad u_0 \gg 1, \quad (14)$$

permitting, in general, to determine the absorption cross sections  $\sigma_0$  and  $\sigma_e$  from the initial and the final parts of the transmission curves. As it is seen from (13), in order to determine the parameter  $\gamma_1$ , it is necessary to use the fitting procedure and to find the limit value  $T_0$  of transmission when  $u_0 \rightarrow 0$ , instead of simply using the experimentally registered minimum transmission value.

However, as we have already mentioned above, experimental possibility to obtain the rectangular distribution of intensity in a sufficiently wide beam is not always available. On the other hand, utilization of small hard diaphragms for separating the rectangular part of the beam results in significant diffraction



distortions of the exciting beam. Therefore, the direct utilization of formulae (13)–(14) again introduces large, unpredictable errors to the results of solution of the inverse problem. Therefore, utilization of sufficiently broad beams with the Gaussian transverse intensity distribution is more reasonable.

Since the publication of [25], it is well known (see also [26] and references therein) that equation (12) with the non-zero linear absorption ( $\alpha \neq 0$ ) does not have analytical solution in the entire region of possible values of the energy density of the exciting radiation. In case of instantaneous relaxation of the excited levels, the presence of the excited-state absorption can be thought of as simply increasing the linear absorption from  $\alpha$  to  $\gamma_2$ . Therefore, only certain approximate solutions of equation (12) are possible. One of these kinds of approximations is proposed in [10] that can be expressed as follows in our notations:

$$T_A = \frac{1 - \exp(-\gamma_2)}{1 - \exp(-\gamma_1)} T_0 + \frac{\exp(-\gamma_2) - \exp(-\gamma_1)}{1 - \exp(-\gamma_1)} T_{FN}, \quad (15)$$

where  $T_{FN} = (\sigma_0 u_0)^{-1} \ln\{1 + \exp(-\gamma_1)[\exp(\sigma_0 u_0) - 1]\}$  is the Frantz-Nodvik transmission in a two-level media approximation. Although this approximate expression deviates from the exact solution of equation (12) quite insignificantly throughout the entire interval of the values of the energy density [13], the approximation for the low energy densities obtained using this expression differs from approximation (13) obtained using the approximation of the initial equation. We shall note that approximate expression (10) in [10] for the transmission coefficient lacks a factor of the form  $1/(1 - T_0)$  in front of the term linear with respect to  $u_0$ . We would also like to draw attention to the fact that the transmission values obtained at  $u_0 \rightarrow 0$  should be used as  $T_0 = \exp(-\gamma_1)$  in the above expressions rather than simply the minimum values of transmission obtained in the experiment [10].

In case of sufficiently broad Gaussian beams  $u_G(r) = u(r, z = 0) = u_0 \exp(-2r^2/w_0^2)$ , equation (12) can be solved independently for different values of the radius  $r$  and then formula (11) used for calculation of the integral transmission. For this type of beams with the energy density  $u_0 \ll 1$  at the beam axis, an approximate expression for the transmission that is convenient for determination of the parameter  $\gamma_1$  can be obtained. Substitution of the expression

$$u(r,1) = \frac{u_G(r)}{a + bu_G(r)} \quad (16)$$

into (11) for the normalized energy density at the exit from the SA and integration over the transverse cross section gives the following expression for the transmitted energy:

$$W(1) = 2\pi \int_0^\infty u(r,1) r dr = \frac{\pi w_0^2}{2b} \ln \left| 1 + \frac{b}{a} u_0 \right|. \quad (17)$$

Using  $\ln(1+x) \approx x - x^2/2$  and  $W(0) = u_0 \pi w_0^2 / 2$ , it is easy to obtain the following expression for the energy transmission coefficient:

$$T(W(0)) \approx \frac{1}{a} - \frac{bW(0)}{\pi w_0^2 a^2}. \quad (18)$$

Since  $b < 0$ , the transmission increases linearly as the normalized energy increases, and the minimum transmission value is given by the limit  $W(0) \rightarrow 0$ . Therefore, for the Gaussian beams, in the same way as for the rectangular ones, the minimal transmission differs from the experimentally measured minimal value and equals  $T_0 = \exp(-\gamma_1)$ . For instance, determination of  $\gamma_1^e$  from the experimentally measured minimum transmission value  $T_{\min}^e = 0.138$  (see Fig. 1) gives  $\gamma_1^e = -\ln(T_{\min}^e) = 1.98$ , while the standard procedure of the linear fitting gives  $T_0 = (13.18 \pm 0.03)\%$  and  $\gamma_1 = 2.03$ . As it will be seen below, this small discrepancy results in deviation of the cross section value  $\sigma_0$  by  $\sim 7.4\%$ .

Thus, using the standard four-level model for modeling the transmission of the sufficiently broad Gaussian beams, only two parameters must be determined:  $\sigma_0$  and  $\gamma_2$ . Independent data about the concentration  $n_0$  of the  $\text{Cr}^{4+}$  ions in the tetrahedral positions usually are not available. The normalized coefficient of the nonresonant absorption  $\alpha$  is not contained explicitly in equation (12). When using it as an independent parameter, the initial density and the excited-state absorption cross section can be expressed as follows:

$$n_0 = (\gamma_1 - \alpha) / \sigma_0, \quad \sigma_e = \sigma_0 (\gamma_2 - \alpha) / (\gamma_1 - \alpha). \quad (19)$$

Therefore, in order to determine  $\sigma_e$ , certain assumptions should be made about the value  $\alpha$ . It is usually assumed that this coefficient is small, and it is set  $\alpha = 0$  in (19). However, in Ref. [14] it is assumed that there is no absorption at all from the excited states of the  $\text{Cr}^{4+}$  ions, i.e.  $\gamma_2 = \alpha$  and  $\sigma_e = 0$ . In the assumption of the instantaneous relaxation of excitations from the upper levels of  $\text{Cr}^{4+}$ , it is indeed impossible to differentiate between the contributions of the excited-state absorption and the linear absorption not only in the processes of the  $\text{Cr}^{4+}$  transmission saturation, but also in case of utilization of the latter for the passive  $Q$ -switching [12, 19-21]. Therefore, in order to conclude the discussion about the presence/absence of the excited-state absorption in  $\text{Cr}^{4+}$ , special experiments are necessary, e.g., the measurements of the photoconductivity, etc.

In order to account for the transverse energy density distribution of the incident collimated pump beam in calculation of transmission, it is necessary to divide the transverse cross section of the beam into thin concentric rings. Solution of equation (12) for each of these rings gives the transmission  $T(u(r))$ . The output energy of the pulse is then given by

$$W(1) = 2\pi \int_0^\infty T(u(r))u(r)rdr . \quad (20)$$

In case of the Gaussian beam, introducing a new integration variable  $q = u_0 \exp(-2r^2 / w_0^2)$ , we obtain the following expression for the transmission coefficient:

$$T_G(u_0) = W(1) / W(0) = (1/u_0) \int_0^{u_0} T(q) dq , \quad (21)$$

where  $T(q)$  is the transmission coefficient calculated in the plane wave approximation. This result has a clear geometrical interpretation (Fig. 2). The integral in (21) gives the area under the transmission curve calculated in the plane wave approximation (solid line). This area is equal to an area under the horizontal dashed line at the level equal to the value of the transmission for the Gaussian beam, i.e., expression (21) can be thought of as a weighted average of the transmission values calculated using the plane wave approximation. For comparison, the dotted line in Fig. 2 denotes the value of the transmission  $T_B = T(u_0 / 2)$  calculated using the plane wave approximation, as proposed in [10]. In Fig. 2 it is seen how the latter approximation overestimates the calculated transmission value for a Gaussian beam.

As stated above, from the three parameters  $\sigma_0$ ,  $\gamma_1$ ,  $\gamma_2$ , the parameter  $\gamma_1$  is easiest to determine in the four level model from the initial part of the transmission data. Determination of the actual values of the other two parameters  $\sigma_0$ ,  $\gamma_2$  from the experimental transmission data is carried out by minimizing the quantity [24,27,28]

$$\chi^2 = \sum_{j=1}^N (T_{c,j} - T_{e,j})^2 / \sigma_T^2 (N-2), \quad (22)$$

that characterizes the difference between the experimental and calculated transmission values. Expression (22) is regarded as a function of two variables  $\sigma_0$ ,  $\gamma_2$ , while the parameter  $\gamma_1$  is determined from the minimum transmission value  $T_0$ . Since the theoretical values of the transmission  $T_{c,j}$  are numerically calculated solving Eq. (12), for minimization of (22) we used the downhill simplex method [24,29], because it does not require calculation of the function

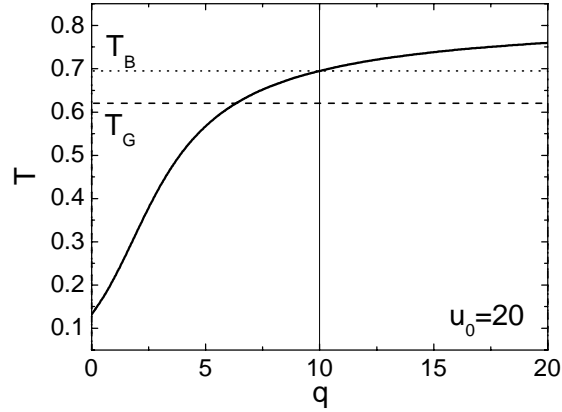


Fig. 2. Geometrical interpretation of the Gaussian beam transmission calculation.

derivatives. If the fitting is correct, one could set  $\chi^2 = 1$  and estimate  $\sigma_T$  via [24,28]

$$\sigma_T^2 = \sum_{j=1}^N (T_{c,j} - T_{e,j})^2 / (N-2). \quad (23)$$

In this way, the required values of the parameters  $\sigma_{0,\min}$  and  $\gamma_{2,\min}$  are determined by minimizing (22) or (23). As it was mentioned above, relatively small errors in determination of the parameter  $\gamma_1$  ( $\Delta\gamma_1/\gamma_1 \sim 2\%$ ) result in a significantly larger deviations ( $\Delta\sigma_{0,\min}/\sigma_{0,\min} \sim 7.4\%$  and  $\Delta\gamma_{2,\min}/\gamma_{2,\min} \sim 3.7\%$ ) from the optimal  $\sigma_{0,\min}$  and  $\gamma_{2,\min}$  that minimize the  $\chi^2$  (22) and the dispersion (23).

Systematic errors in measurement of the beam energy and radius, as it is seen from (18), practically do not influence the value of  $\gamma_1$ , since it is determined at  $W(0) \rightarrow 0$ . At the same time, systematic errors in determination of the energy density on the beam axis arising from the corresponding errors in measurements of the total energy or the beam radius lead to a shift of the values of  $\sigma_{0,\min}$  and  $\gamma_{2,\min}$ . It can be verified easily by artificially multiplying the energy densities of the experimental data by an appropriate factor, while leaving the transmission values at the corresponding points the same, thereby simulating a systematic error in experimental measurement of the former, and then determining the absorption cross-sections from the nonlinear fitting of the curve with modified energy densities. E.g., the error of 4% in measurement of the beam radius result in an error of up to  $\sim 8.5\%$  in determination of the ground-state absorption cross section. In general, it follows from the calculations that the systematic error in measurement of the axial energy density  $|\Delta u_0/u_0|$  results in a relative shift  $|\Delta\sigma_{0,\min}/\sigma_{0,\min}|$  of the same absolute value and the opposite sign.

Rough errors in normalizing the experimental data or converting the energy densities to dimensionless units result in obtaining the false values of  $\sigma_{0,\min}$  and  $\gamma_{2,\min}$  that are far from the real values of the absorption cross-sections. However, it is difficult to detect the presence of serious errors without additional research, e.g., analysis of generation dynamics of lasers with passive  $Q$ -switching [20,21], since the dispersion  $\sigma_T$  does not change drastically enough in that case.

We shall note that the physics papers usually do not inform what considerations are used as the basis for indication of the certain measurement errors. As the basis for assessment of these errors, the above mentioned shifts  $\sigma_{0,\min}$  and  $\gamma_{2,\min}$  arising on the boundaries of the interval of the standard deviation in the measurement of the axial energy density can be used. However, these mean square deviations of the axial energy density usually do not exceed 10%. We shall demonstrate that the procedure of their determination using the

least squares method leads to considerably larger uncertainty intervals of the parameters being determined.

The confidence limits of the parameter values given by the minimization procedure can be assessed by calculating the deviation of the parameters  $\Delta\sigma_0$  and  $\Delta\gamma_2$  from the optimal values  $\sigma_{0,\min}$ ,  $\gamma_{2,\min}$  giving the constant values of  $\chi^2 = \chi_{\min}^2 + \Delta\chi^2$  with  $\Delta\chi^2 = 1.0, 2.71$  and  $6.63$  [24]. Fig. 3 depicts the isolines

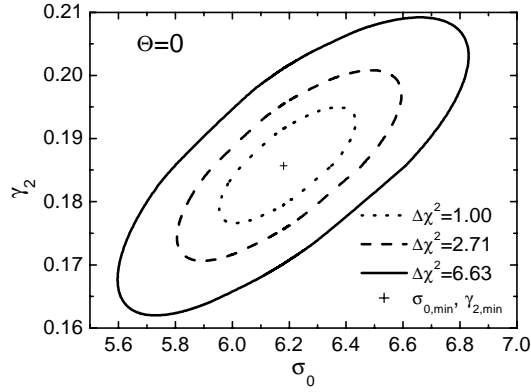


Fig. 3. Confidence region “ellipses” corresponding to values of chi-square larger than the fitted minimum.

of the constant values of  $\chi^2$ . The confidence limits of a fitting parameter are then given by the projection of the corresponding ellipse to the axis of this parameter, e.g., the interval given by the projection of the ellipse with  $\Delta\chi^2 = 6.63$  contains 99% of the normally distributed data [24].

Determination of the absorption cross-sections from the transmission data for the SA rotated by 45 degrees (the lower set of points in Fig. 1) results in relatively small shift of the mean values  $\tilde{\sigma}_{0,\min}$ ,  $\tilde{\gamma}_{2,\min}$  minimizing (22), (23) and increase of the confidence intervals.

#### 4 Conclusion

We have shown that for precise description of the energy transmission and determination of absorption cross sections of the  $\text{Cr}^{4+}$ :YAG crystals, it is sufficient to use the standard four-level scheme with fast relaxation excitation, taking into account the anisotropy of absorption and nonhomogeneous

transverse energy distribution of nano- and subnanosecond pump pulses. Critical analysis of typical simplifications commonly used for solving the inverse problem of determination of absorption cross sections is undertaken. A simple technique for determination of confidence intervals in case of the nonlinear regression is emphasized.

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