

Study of the uniqueness and stability of the solution of inverse problem in saturation fluorimetry

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Abstract. The uniqueness and stability of the solutions of two- and three-parameter inverse problems in nonlinear fluorimetry (saturation fluorimetry) of complex organic compounds is studied by the method of computer simulations. The main attention is paid to a practical stability of such problems with respect to the input data noise and variations in the model. The use of the technique of artificial neural networks (including their training on noisy input data) provides the solution stability under real experimental conditions.

1. Introduction

This work was initiated by studies in the field of laser diagnostics of complex organic compounds (COCs) in aqueous media [1, 2]. At the same time, the problem of the uniqueness and stability of the inverse problems of nonlinear laser fluorimetry (saturation fluorimetry) is of general interest. Fluorimetry is extensively used for diagnostics of organic compounds in aqueous media [1–4], although it faces a number of obstacles, in particular, the problem of COC identification. In some cases, this problem cannot be solved within the framework of a phenomenological approach only, i.e., using only fluorescence and fluorescence excitation spectra and even total luminescence spectra, although these spectra provide extensive information.

The above statement can be illustrated by the fluorescence spectra of humus, which is contained in some concentration in natural water [3], tap water, and even (in trace amounts) in purified technological water used in microelectronics. Although the humic substance composition is different for different media (in particular, the ratio of humic and fulvic acids changes), its fluorescence and fluorescence excitation spectra, which represent broad structureless bands, are virtually indistinguishable [3], which severely restricts the possibilities of conventional ('linear') fluorimetry for diagnostics of this important component of aqueous media.

Similar obstacles are often encountered in fluorescence *in situ* diagnostics of petroleum pollutants of aqueous media,

especially when their fluorescence is weaker than that of humic substance. One of the ways to increase the information content of fluorimetry is to use a nonlinear regime. In this case, along with conventional fluorescent parameters (position and shape of the fluorescence band, the fluorescence decay time, etc.), one determines molecular photophysical parameters of fluorophores such as the absorption, fluorescence, and fluorescence excitation cross sections, and the rate constants of intramolecular transitions and intermolecular energy transfer which can be used in diagnostics of organic impurities [1–4].

It is important to emphasise that in this way not only applied problems of diagnostics of organic complexes are solved but also fundamental problems. The mechanisms of photophysical molecular processes proceeding upon interaction of laser radiation with COCs are established, the nature of fluorescence bands is elucidated, and genesis and transformation of COCs are studied. All these parameters should be measured *in vivo*. Such diagnostics, which is based on the measurement of photophysical parameters of COCs can be performed by means of nonlinear fluorimetry [1, 2, 4–6].

Fluorescence saturation upon powerful laser excitation [7] represents a nonlinear dependence of the number N_{fl} of fluorescence photons on the photon flux density F of exciting radiation (Fig. 1). The fluorescence saturation curves can be plotted in different forms: $N_{\text{fl}}(F)$, $\Gamma(F)$ etc., where Γ is the fluorescence saturation factor, which is determined from the expression

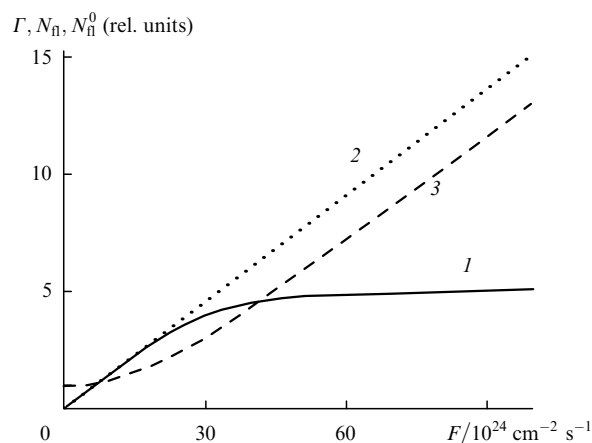


Figure 1. Dependences of the number of fluorescence photons N_{fl} (1) and N_{fl}^0 (2) and the saturation factor Γ (3) on the exciting radiation flux density.

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$$\Gamma(F) = \frac{N_{\text{fl}}^0}{N_{\text{fl}}};$$

where N_{fl} is a real number of fluorescence photons; N_{fl}^0 is the number of fluorescence photons that would be emitted in the absence of saturation, i.e., in the case of a linear dependence of N_{fl} on N_{fl}^0 (Fig. 1).

Fluorescence of organic molecules can be saturated for several reasons such as the dynamic depletion of the ground state of the molecules, intersystem crossing, absorption from excited states, intermolecular interactions of the singlet-singlet (ss) annihilation type, etc. It is obvious that these parameters of the saturation curve are determined by photophysical characteristics of the molecule and, hence, these characteristics can be found from the saturation curves. The solution of this inverse problem is used as the basis for nonlinear fluorimetry applied to the spectral analysis of fluorescing atoms and molecules.

2. Inverse problems of saturation fluorimetry

As noted above, the inverse problem of saturation fluorimetry consists in the determination of molecular photophysical parameters from the fluorescence response (the saturation curve) within the framework of the model applied. The mathematical formulation of such a problem was given in Ref. [8], where the problem was shown to be correct according to Tikhonov [9], i.e., its solution is unique and stable on some chosen set of solutions (the authors of Ref. [8] meant theoretical uniqueness and stability of the solution).

The photophysical model that we used in the inverse problem of saturation fluorimetry involved the following processes: absorption of light, radiative and nonradiative relaxation, intersystem crossing, and intermolecular interactions [8]. We neglected transitions from the triplet state to the ground singlet state by restricting our consideration to the problems in which the duration of the exciting laser pulse $t_p \approx 10^{-8}$ s, i.e., is substantially shorter than the triplet state lifetime.

We also ignored absorption of the pump radiation and fluorescence by molecules in the excited singlet (S_1) and triplet (T_1) states and multiphoton absorption, although some of these processes (but not all and not always) can affect fluorescence saturation curves, which requires a special consideration during the choice of an object and conditions of the experiment.

In accordance with the chosen model of photophysical processes in organic molecules, we can write the system of kinetic equations for populations n_1 , n_2 , and n_3 of the energy states S_0 , T_1 , and S_1 :

$$\begin{aligned} \frac{\partial n_1(t, \mathbf{r})}{\partial t} &= -F(t, \mathbf{r})\sigma_{13}n_1(t, \mathbf{r}) + (K_{31} + K'_{31})n_3(t, \mathbf{r}) + \gamma n_3^2(t, \mathbf{r}), \\ \frac{\partial n_2(t, \mathbf{r})}{\partial t} &= K_{32}n_3(t, \mathbf{r}), \\ \frac{\partial n_3(t, \mathbf{r})}{\partial t} &= F(t, \mathbf{r})\sigma_{13}n_1(t, \mathbf{r}) - (K_{31} + K'_{31})n_3(t, \mathbf{r}) \\ &\quad - K_{32}n_3(t, \mathbf{r}) - \gamma n_3^2(t, \mathbf{r}), \end{aligned} \quad (1)$$

$$n_1 + n_2 + n_3 = n_0,$$

where $F(t, \mathbf{r}) = F_0 f(t/t_p)g(r/r_0)$ is the photon flux density of exciting radiation; r_0 is the laser beam radius; $f(t/t_p)$ and $g(r/r_0)$ are temporal and spatial distributions of laser radiation, respectively; $\mathbf{r} = (x, y)$ and z are co-ordinates directed perpendicular and parallel to the laser beam, respectively; σ_{13} is the absorption cross section for exciting radiation; $K_3 = K_{31} + K'_{31} + K_{32}$; K_{31} and K'_{31} are the rates of radiative and nonradiative transitions from the S_1 state to the S_0 state; K_{23} is the rate of the $S_1 \rightarrow T_1$ transition; γ is the rate constant of the ss annihilation; n_0 is the concentration of fluorescing molecules; γn_3 is the rate of the ss annihilation; and γn_0 is the maximum rate of the ss annihilation.

By solving the system of equations (1) analytically or numerically, we can find the population n_3 of the first excited level and the number N_{fl} of fluorescence photons emitted from the volume $V = Sl$ (S is the beam cross section and l is the thickness of the layer from which fluorescence is detected):

$$N_{\text{fl}} = K_{31} \int_0^\infty dt \int_S d\mathbf{r} \int_0^l dz n_3(t, \mathbf{r}, z). \quad (2)$$

Using relations (1) and (2), we can obtain theoretical curves of the fluorescence saturation, which are required for solution of the inverse problem.

One can see from (1) and (2) that the fluorescence saturation curve depends on the following photophysical parameters: (i) the absorption cross section σ_{13} ; (ii) the S_1 state lifetime $\tau_3 = K_3^{-1}$; (iii) the quantum yield of molecules to the lower triplet state $S_1 \rightarrow T_1$ $\eta_T = K_{32}/K_3$; and (iv) the rate constant of the ss annihilation γ . We assume that temporal, $f(t/t_p)$, and spatial, $g(r/r_0)$, distributions of laser radiation, which also affect the saturation curve, are known. Therefore, our model of photophysical processes leads to the four-parameter inverse problem of saturation fluorimetry whose solutions are parameters σ_{13} , τ_3 , η_T and γn_0 .

At present, as our earlier studies showed [6], the satisfactory accuracy (for real experimental errors of the input data) can be obtained for one-, two-, and three-parameter problems. We will analyse the uniqueness and stability of solutions namely for these problems. One can expect that the dimensionality of solvable inverse problems will be increased in the future. However, the approach based on reducing the problem dimensionality by different methods (introduction of generalised parameters, successive solution of the problems with artificially reduced dimensionality, etc.) is more efficient.

An important step in the solution of inverse problems is the proof of the uniqueness and stability of solutions. One should study both theoretical and practical uniqueness and stability. The theoretically stable solution can become unstable when the input data noticeably deviate from the exact model dependence. In this case, the solution can considerably differ from the real solution. Such instability is called practical because it can appear in real (and numerical) experiments.

The input data may deviate from the model fluorescence saturation curve for two reasons: incorrect choice of the model of photophysical processes (for example, neglect of some physical processes in molecules, incorrect values of fixed parameters, including distributions $f(t/t_p)$ and $g(r/r_0)$, etc.) and large noise of the input data (i.e., a large error of the saturation curve measurement).

It is reasonable to introduce a concept of the threshold of practical instability – the deviation of the input data at which the solution strongly differs from the solution of an ‘ideal’ theoretical problem. As a criterion for practical instability, we chose the deviation of the solution exceeding the specified range of variation of required parameters.

The practical uniqueness means that the input data measured with some error correspond to a unique set of the required parameters that satisfies to the practical stability criterion. If the problem is practically unstable, its solution is obviously not unique or does not exist. Thus, the practical stability is a key question. This question is especially important in inverse problems of nonlinear fluorimetry of COCs because the specific feature and complexity of these problems is that the saturation curves are smooth, without any extrema, inflections, etc., and they are weakly ‘sensitive’ to photophysical parameters [2, 6].

Note also that the practical stability substantially depends on the solution algorithm of the inverse problem. The solutions of inverse problems of saturation fluorimetry of COCs obtained with the help of various algorithms [2, 5, 6, 10] suggest that the best practical stability is provided by algorithms based on the method of artificial neural networks (ANN) [11]. Here, we studied the practical stability of a solution of the inverse problem of saturation fluorimetry with the help of ANN.

We solved our problem using a three-layer perceptron [11] with 16 neurones in a hidden layer. The ANN was trained using model saturation curves, which were calculated for the following photophysical parameters: $\sigma_{13} = 10^{-17} - 10^{-16}$ cm², $\tau_3 = 1 - 10$ ns, $\eta_T = 10^{-2} - 10^{-1}$, $\gamma_{n0} = 10^7 - 10^9$ s⁻¹. These values of parameters are typical for solutions of dyes and related compounds at concentrations between 10^{-5} and 10^{-3} M.

We used two variants of neural network training by employing the saturation curves without noise and noisy curves. In the first case [6], the neural network was trained and tested by presenting theoretical ‘ideal’ saturation curves. As shown in Ref. [6], the presentation of the curves with experimental errors resulted in the development of practical instability already at low noise. For this reason, we used the noisy input data for the ANN training by presenting the noisy saturation curves to the network along with ideal curves. The saturation curves were made noisy using a random-number generator with a uniform distribution. We specified the maximum noise, and to the network the curves were presented with the noise amplitude chosen randomly from zero to maximum.

The criterion of the network quality and the root-mean-square error of the reconstruction of parameters is the coefficient of multiple determination

$$R^2 = 1 - \frac{\sum_i (y_i - \tilde{y}_i)^2}{\sum_i (y_i - \bar{y})^2}, \quad (3)$$

where $y_i, \tilde{y}_i, \bar{y}$ are true, predicted, and mean values of the parameter being determined in the chosen range of its variation, respectively.

We observed a distinct correlation between the coefficient R^2 and the root-mean-square error of reconstruction of parameters: the fall of R^2 to zero approximately corresponded to root-mean-square errors of reconstruction of parameters

comparable to the range of their variation. This allowed us to use the parameter R^2 as a factor of practical stability of the solution of the problem by this method and consider the equality $R^2 = 0$ as a threshold of practical instability of the inverse problem solution.

3. Uniqueness of a solution of the inverse problem of saturation fluorimetry

The uniqueness of a solution of the inverse problem under study has been rigorously proved in Ref. [8] for the saturation curve represented in the form $N_{fl}(F)$ and for a rectangular space-time distribution of the exciting radiation intensity. Although we did not prove the uniqueness of the representation for $\Gamma(F)$, we can assume in a qualitative comparison of our situation with the problem [8] the uniqueness of solutions in the cases when the saturation factor $\Gamma(F)$ can be obtained analytically. Unfortunately, in the case of an arbitrary space-time distribution of the exciting radiation intensity it is impossible to obtain an analytic expression for $\Gamma(F)$. For this reason, we verified the uniqueness of the reconstruction of three parameters ($\sigma_{13}, \tau_3, \gamma_{n0}$) in the case of an arbitrary time distribution and a rectangular spatial distribution by numerically solving the inverse problem by the ANN method.

We presented three saturation curves, which differed at each point no more than by 0.5% (i.e., were virtually coincident by eye), to the neural network trained on the curves without noise. These curves were calculated for different sets of parameters $\{\alpha, \sigma_{13}, \gamma_{n0}\}$, where $\alpha = \tau_3/t_p$ (true parameters in Table 1). The results of presentation of the saturation curves to the neural network are given in rows with noise 0 and 1% in Table 1. One can see that the network distinguished these curves (both without noise and with the 1% noise) and reconstructed photophysical parameters with an error of no more than 19% (see the ‘root-mean-square deviation’ column), which is substantially lower than the difference in the specified input data. Note that an extremely small difference between the saturation curves for the sets of apparently strongly different parameters is explained by the fact that the product $\tau_3\sigma_{13}$ is the same for all the three cases, while the parameter γ_{n0} only slightly exceeds the parameter τ_3^{-1} and, hence, the acting (rather than maximum) rates γ_{n3} of the ss-annihilation are lower than the rate τ_3^{-1} .

This example illustrates (but does not prove rigorously) the theoretical uniqueness of the inverse problem solution. In addition, the obtained results demonstrate a high efficiency of the ANN method for solving inverse problems. We demonstrated once more the unique property of ANN – their ability to learn by examples and not simply remember but generalise the input data and to reveal the hidden regularities in order to recognise the patterns.

4. Stability of a solution of the inverse problem of saturation fluorimetry

As noted above, the real fluorescence saturation curves differ from the calculated ‘model’ curves because of the random measurement errors and possible deviations of photophysical processes from the model used. In numerical experiments, noise is simulated by a random-number generator, which generates the numbers uniformly in a specified range (the noise amplitude). A set of varied and fixed parameters and parameters of the exciting radiation will be called the model.

Table 1. Simultaneous determination of three photophysical parameters from model saturation curves by the ANN method.

Number of the saturation curve	Noise (%)	τ_3/ns	Root-mean-square deviation (%)	σ_{13}/cm^2	Root-mean-square deviation (%)	$\gamma m_0/\text{s}^{-1}$	Root-mean-square deviation (%)
1	True	10		$2.28 \cdot 10^{-17}$		$2.97 \cdot 10^8$	
	0	9.7	11.9	$1.7 \cdot 10^{-17}$	15	$2.31 \cdot 10^8$	12.27
	1	8.8	17.6	$2.31 \cdot 10^{-17}$	18.6	$2.64 \cdot 10^8$	14
2	True	6		$3.8 \cdot 10^{-17}$		$3.8 \cdot 10^8$	
	0	6.3	11.9	$4.2 \cdot 10^{-17}$	15	$4.4 \cdot 10^8$	12.27
	1		17.6	$2.28 \cdot 10^{-17}$	18.6	$2.97 \cdot 10^8$	14
3	True	4		$5.7 \cdot 10^{-17}$		$4.88 \cdot 10^8$	
	0	3.9	11.9	$6.4 \cdot 10^{-17}$	15	$5.9 \cdot 10^8$	12.27
	1	3.4	17.6	$6.8 \cdot 10^{-17}$	18.6	$6 \cdot 10^8$	14

We will study the solution stability with respect to the model subjected to different variations.

4.1. Stability of a solution of the inverse problem of nonlinear fluorimetry with respect to the input data noise

Using the ANN technique, we solved two two-parameter ($\{\sigma_{13}, \tau_3, K_{32} = 5 \cdot 10^8 \text{ s}^{-1}, \gamma m_0 = 0\}$ and $\{\sigma_{13}, \tau_3, K_{32} = 5 \cdot 10^7 \text{ s}^{-1}, \gamma m_0 = 5 \cdot 10^8 \text{ s}^{-1}\}$) and two three-parameter ($\{\sigma_{13}, \tau_3, \eta_T, \gamma m_0 = 0\}$ and $\{\sigma_{13}, \tau_3, \eta_T = 0, \gamma m_0\}$) problems. The curves were calculated using the following distributions of the laser pulse in time and space

$$f(\tau) = \frac{1}{\ln 2} \frac{\tanh(2\tau)}{\cosh^2 \tau}, \quad g(r) = \left(\frac{4 \ln 2}{\pi} \right)^{1/2} \exp[-(4 \ln 2)\rho^2],$$

where $\tau = t/t_p$; $\rho = r/r_0$.

The calculations showed that the threshold of practical instability of the solutions of two-parameter problems upon the ANN training on the noiseless curves is achieved for the 10-% input data noise for both parameters. The threshold of practical instability for the three-parameter problems is noticeably lower, and for the parameter σ_{13} it is achieved when the input data noise for two other parameters is 1 and 3%. These results are illustrated in Fig. 2.

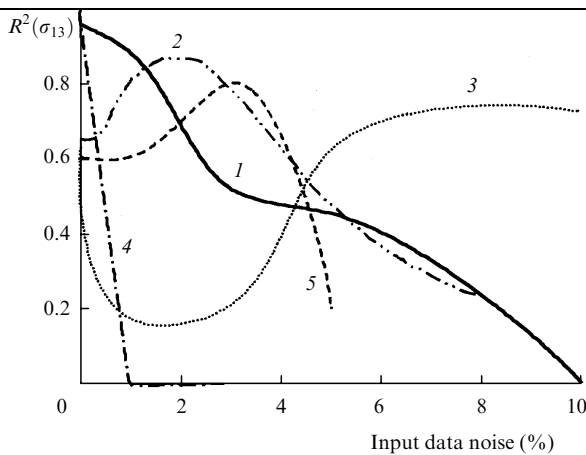


Figure 2. Dependences of coefficients of multiple determination (for the parameter σ_{13}) on the noise of fluorescence saturation curves for two-parameter $\{\sigma_{13}, \tau_3, K_{32} = 5 \cdot 10^7 \text{ s}^{-1}, \gamma m_0 = 0\}$ (1–3) and three-parameter $\{\sigma_{13}, \tau_3, \eta_T, \gamma m_0 = 0\}$ (4, 5) problems. The noise upon training was 0 (1, 4), 10 (2), and 50% (3, 5).

The ANN technique proves to be more efficient if the neural network is trained on the noisy fluorescence saturation curves. The neural networks were trained for each of the two- and three-parametric problems for the maximum noise of the training input data equal to 10, 50, and 100%. Some of the results (with the 10- and 50-% input data noise) are presented in Fig. 2. One can see that training on noisy data substantially increases the instability threshold for the solutions of inverse problems.

The obtained results distinctly demonstrate that when the input data noise during the neural network training is large, the parameters are better reconstructed from the noisy saturation curves than from the noiseless curves. In this case, the coefficient of multiple determination does not decrease monotonically but has a maximum at some noise of the presented saturation curves. The input data noise at which the coefficient R^2 has a maximum value increases with increasing training noise.

One can see from Fig. 2 that the ANN training on noisy curves allows one to increase the stability threshold for the solutions of inverse problems with respect to the input data noise in two- and three-parameter problems up to ~ 15 and 5%, respectively, which corresponds to typical errors of the real experiment. Therefore, if the preliminary information on the input data error is available, the neural network training optimised to this noise is possible.

4.2. Stability of a solution of the inverse problem of nonlinear fluorimetry with respect to variations in the model

We considered first the practical stability of the problem solution with respect to variation in one of the model parameters. We solved the following two-parameter problems: $\{\sigma_{13} = \text{const}, \tau_3, \eta_T, \gamma m_0 = 0\}$, $\{\sigma_{13}, \tau_3 = \text{const}, \eta_T, \gamma m_0 = 0\}$, $\{\sigma_{13}, \tau_3, \eta_T = \text{const}, \gamma m_0 = 0\}$, $\{\sigma_{13}, \tau_3, \eta_T = 0, \gamma m_0 = \text{const}\}$.

We solved these problems using the ANN technique by training networks on the noiseless input data. Then, we calculated the sets of fluorescence saturation curves in which the value of the fixed parameter was successively varied within its entire range. These sets with noises 0.1 and 3% were presented to the trained networks. Fig. 3 shows some of the dependences of the coefficient of multiple determination R^2 on the parameter determining the model (which takes fixed values in our case).

These dependences represent resonance curves with a maximum at the same value of the parameter for which

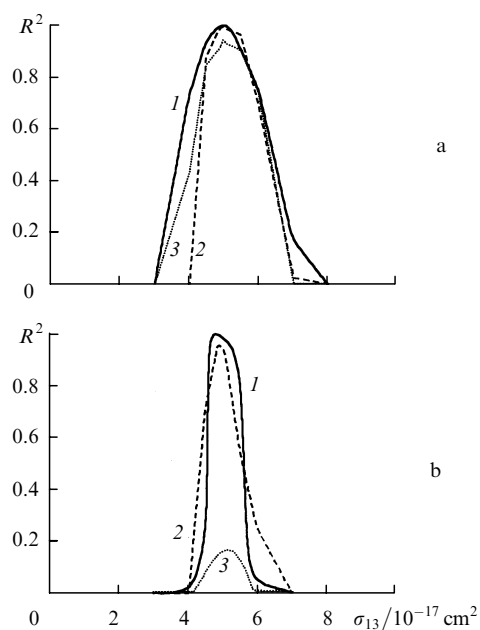


Figure 3. Dependences of the coefficient of multiple determination R^2 for τ_3 (a) and K_{32} (b) on the parameter σ_{13} determining the model of photophysical processes for the neural network trained for $\sigma_{13} = 5 \cdot 10^{-17} \text{ cm}^2$ and the input data noise equal to 0 (1), 1 (2), and 3% (3).

the network was trained. We found that when σ_{13} and τ_3 were fixed parameters of the model, the practical instability of the solution appeared when these parameters differed from the true ones by 30%. In models defined by parameters η_T and γn_0 , the practical instability of the solution appeared at substantially greater deviations of these parameters from their values for which the network was trained. These results should be expected, because in the ranges of variation of σ_{13} , τ_3 , η_T and γn_0 chosen by us, the parameters σ_{13} and τ_3 affect the shape of saturation curves in a greater degree than η_T and γn_0 .

Unfortunately, the coefficient of multiple determination R^2 can be determined only in numerical experiments, when many curves with known parameters can be calculated. However, the study of the stability of the inverse problem solution with respect to variations in the model can be used for a correct choice of the model of photophysical processes in real experiments. If the experimental fluorescence saturation curve of some organic compound is presented to a set of neural networks that were trained for various models (with different numbers of variable and fixed parameters), then, because of the resonance properties of the solution, the network that was trained for the model most adequately describing the physical processes proceeding in this organic compound will give the more reasonable response (more reasonable parameters).

Another way to change the model is to increase the number of parameters being determined. First, we considered a two-parameter inverse problem $\{\sigma_{13}, \tau_3, \eta_T = 0, \gamma n_0 = 0\}$. Then, we added to this model either parameter η_T or γn_0 as unknown parameters. From the physical point of view, the consideration of a new parameter should not effect the initial parameters and the accuracy of their measurement. After the addition of the quantum yield η_T to the triplet state of the rate γn_0 of the ss-annihilation, the criterion of the solution stability with respect to the model variation is the invariability of parameters σ_{13} and τ_3 and the accuracy of

Table 2. Root-mean-square errors of measurements of parameters σ_{13} , τ_3 and η_T in solution of two- and three-parameter problems using ANNs trained on the noiseless input data.

Noise (%)	Two-parameter problem		Three-parameter problem		
	$\delta\sigma_{13}$ (%)	$\delta\tau_3$ (%)	$\delta\sigma_{13}$ (%)	$\delta\tau_3$ (%)	$\delta\eta_T$ (%)
0	13	14	12.3	16	18
1	13	18	13	19	21
3	14.8	20	–	21	–

their reconstruction determined from the two-parameter model.

One can see from the results presented in Table 2 that the quality of the reconstruction of parameters σ_{13} and τ_3 in the three-parameter problem (after an addition of η_T) remained quite high. This shows that solutions of inverse problems of saturation fluorimetry are stable to the model variations caused by the addition of parameters.

5. Conclusions

Analysis of the study of the uniqueness and stability of solutions of inverse problems of saturation fluorimetry of organic compounds showed that modern mathematical methods allow one to determine photophysical parameters from saturation curves with sufficiently high accuracy. Note that two or three (possibly, even more in the future) photophysical parameters, which have been conventionally measured with different instruments, are determined simultaneously by the same method using only a laser spectrometer for saturation fluorimetry. Therefore, saturation fluorimetry represents a fundamentally new approach to the fluorescence diagnostics of complex organic compounds, which opens up new possibilities in the solution of applied problems in diagnostics of organic complexes and of many fundamental problems concerning the mechanisms of photophysical processes. The results obtained in this paper confirm once more the known postulate of the theory of inverse problems [9] according to which the successful solution of these problems substantially depends on the volume of *a priori* information.

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