ON SOME ALGORITHMS FOR COMPARING MODELS OF FEMTOSECOND LASER RADIATION PROPAGATION IN A MEDIUM WITH GOLD NANORODS

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Abstract

The study of the features of the nonlinear optical interaction of laser radiation with various media is currently widely studied, including by mathematical modeling methods. In particular, the interaction of the laser pulse with the medium doped with nanorods leads to the melting of these nanorods and distortion of the laser pulse. In addition, the passage of a laser pulse through a medium with nanoparticles can lead to a change in the shape of the pulse and a significant distortion of its spectrum, as a result of which a number of undesirable effects may occur.

The corresponding process can be described by the nonlinear Schrödinger equation. Analytical studies of the corresponding variants of the equation and numerical calculations have shown that the propagation of the resulting soliton subimpulses is characterized by the formation of a so-called nonlinear frequency chirp in the region of subimpulses.

When considering the graphs of such a process, the following assumption arose: it is possible to measure the degree of nonlinearity by calculating the correlation coefficient between the frequency distribution in a linear and nonlinear medium. In the course of calculations using correlation analysis, we obtained statistical characteristics similar to those that were assumed in advance. At the same time, the most adequate variant of the pair correlation coefficient is obtained in the case of applying the algorithm proposed by us for its calculation.

Key words

Femtosecond laser radiation, frequency chirp distributions, mathematical model, rank correlation.

1 Introduction and motivation

In this paper, we consider some algorithms for mathematical modelling femtosecond laser radiation propagation in a medium with gold nanorods, as well as special statistical studies of the calculations obtained in these models.

The study of the features of the nonlinear optical interaction of laser radiation with various media is currently widely studied, including by mathematical modeling methods; they are the subject of this paper. The manifestation of nonlinearity is largely due to the intensity of the incident radiation. The nonlinearity of the interaction may also be due to inclusions of metallic (gold or silver) nanoparticles. Under the action of laser radiation, as a result of the excitation of surface waves (plasmon polaritons), metal nanoparticles can be heated to melting temperatures. As a result of melting, the shape of nanoparticles changes, which in turn leads to a change in the conditions and parameters of the interaction of laser radiation with a nonlinear medium. Optical nonlinear media containing inclusions of metallic nanoparticles are widely used in various applications [Kim et al., 2008; Driben et al., 2009; Patwari and Bheemaiah, 2020; Salih et al., 2021; Zhu et al., 2021], among which five-dimensional optical memory can be highlighted [Zijlstra et al., 2009; Park et al., 2020]. In addition to the three physical dimensions, the recording and reading of information in five-dimensional memory devices also depends on the orientation of the nanoparticles and the ratio of their sides.

When mathematically describing the interaction of laser radiation with optical media containing metallic

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nanoparticles, in addition to equations regarding the electric field strength, it is necessary to write additional equations describing the process of changing the shape of nanoparticles. In [Trofimov and Lysak, 2016a], a mathematical model was proposed that allows taking into account changes in the parameters of nonlinear interaction as a result of changes in the shape of cylindrical nanoparticles. The model is based on the cubic nonlinear Schrödinger equation, the parameters of which depend on the aspect ratio of nanoparticles, and an equation describing the change in the aspect ratio under the action of laser radiation of a certain polarization and frequency as a result of its resonant absorption.

The passage of a laser pulse through a medium with nanoparticles can lead to a change in the shape of the pulse and a significant distortion of its spectrum. As a result, a number of undesirable effects may occur, including false reading and writing of information. It is possible to avoid these undesirable effects by creating conditions for soliton or self-similar pulse propagation. These conditions were studied in [Trofimov and Lysak, 2016a; Trofimov and Lysak, 2017; Trofimov and Lysak, 2018], in which, for the cases of one-, two- and multi-photon resonances, the phenomena of acceleration and deceleration of laser pulses while maintaining their soliton shape, as well as splitting of the incident pulse into several subimpulses of a soliton shape, were demonstrated. Analytical studies and numerical calculations have shown that the propagation of such soliton subimpulses is characterized by the formation of a nonlinear frequency chirp (nonlinear dependence of instantaneous frequency on time) in the subimpulse region. When a pulse propagates in a linear medium, in the absence of interaction of laser radiation with nanoparticles, the frequency chirp has a linear character. Thus, the degree of deviation of the nonlinear frequency chirp from the linear one can serve as an indicator of the nonlinear nature of the interaction of laser radiation with an optical medium containing metal nanoparticles.

Thus, from the point of view of nonlinear optics, the subject of the work is as follows. We applied the algorithm to the problem of femtosecond laser radiation propagation in a medium with golden nanorods. This problem is very important, for example, for 5-dimensional memory, see [Zijlstra et al., 2009; Salih et al., 2021; Zhang et al., 2024] etc. The interaction of a laser pulse with a medium doped with nanorods results in the nanorods melting and the laser pulse distortions. Within the framework of a slowly varying envelope, the process of femtosecond laser pulse propagation in the medium with golden nanorods can be described by a nonlinear Schrödinger equation supplemented with an ordinary differential equation for nanorods aspect ratio, [Driben et al., 2009].

By examining the graphs of the relevant indicators, exactly for the femtosecond laser radiation propagation in a medium doped with golden nanorods and in a linear medium, the following assumption arose.

> *It is possible to measure the degree of nonlinearity by calculating a correlation coefficient between the chirp distribution in a linear and nonlinear medium.*

Then in the rest of the paper, we shall consider the correlation between two corresponding graphs in different ways, and try to draw conclusions that are important for the subject area under consideration. At the same time, we shall focus on various pair correlation algorithms, including the new algorithm proposed in this paper.

From the point of view of mathematical statistics, we can describe motivation as follows. Previously, *in other subject areas*, we tested completely different hypotheses, but we performed such checks in the same way that we describe in this paper. At the same time, we have always received good results, and here the word “good” means here “expected”. We specifically note that the most adequate results were shown by our proposed variant of calculating the pair correlation; this variant is also described in this paper.

The paper has the following structure.

In Section 2, we list the standard ways to calculate the rank correlation. Then in Section 3, we propose our own version for such a calculation; it differs significantly from Spearman’s and Kendall’s versions.

In Section 4, we describe our task in more detail than in Introduction from the point of view of nonlinear optics; at the same time, we pose the problem of why the observed and predicted results should be investigated using correlation analysis.

In Section 5, we give some results of computing experiments for the real data previously given in Section 4. Certainly, Section 5 is the main section of the paper, despite its small volume.

Section 6 is the conclusion: we formulate some direction for the future work.

2 On the rank correlation: various approaches

In this section, we list the standard ways to calculate the rank correlation.

Thus, let us consider some usual statistical characteristics used in the paper, are agreed with [Lagutin, 2012; Wasserman, 2013]. Sometimes, we use “some more mathematical” notation, for example, we do not use MXY etc. The two random variables under consideration are denoted by X and Y; their observed implementations are denoted in the same way with the corresponding subscripts, i.e.,

$$X_i \text{ and } Y_i \text{ for } i = 1, 2, \ldots, N.$$

Firstly, let us formulate *the usual definition of correlation*: recall that the pair correlation coefficient can be calculated using the usual formulas:

$$R(X, Y) = \frac{\operatorname{cov}(X, Y)}{\sigma_X \cdot \sigma_Y},$$

where

$$\mathrm{cov}(X, Y) = M_{X \cdot Y} - M_X \cdot M_Y \,.$$

In our further tables, this variant of the coefficient *will have the number* 0.

Secondly, let us formulate *some modificated Kendall's correlation coefficient*. For it, we define *the number of discrepancies* ("entropy coefficient"): a discrepancy holds if for some pair (i, j) where $i \neq j$, we have

$$X_i > X_j \;\text{ but }\; Y_i < Y_j \,. \tag{1}$$

In the next formulae, let us denote the number of such discrepancies by $entr(X, Y)$, or simple E if the random variables are implied.

Since the maximum possible number of such discrepancies is $\frac{N \cdot (N-1)}{2}$, we shall consider the modificated Kendall's correlation coefficient by

$$1 - \frac{4 \cdot E}{N \cdot (N-1)};$$

this value is equal to 1 in case of 0 discrepancies, and is equal to -1 in case of maximum possible number of discrepancies. In our further tables and program fragments (Fig. 1), this variant of the coefficient *will have the number* 2.

Note that we could calculate this coefficient as follows. We define the "entropy coefficient" considered before for each pair of pairs by (1), then we calculate the sum of these coefficients and divide the result by the value $\frac{N \cdot (N-1)}{2}$ already used earlier.

However, different publications provide different versions of criticism of the Kendall criterion, but the authors of the current paper consider such a flaw to be the most important: it does not give very adequate results with a large number of coincidences in the values of the considered random variables. Therefore we shall also consider the following *"very modificated" Kendall's correlation coefficient*.

It is most convenient to consider it as a search for pairs of pairs, like in the last remark. However, unlike (1), we also use values 0 (not only 1 and -1): the value 0 is selected if and only if the values of at least one of the random variables in the considered pairs match.

In our further tables and program fragments (Fig. 1), this variant of the coefficient *will have the number* 3.

Thirdly, the *Spearman's correlation coefficient* is calculated in the usual way, i.e.

$$\frac{\sum\limits_{i=1}^{n} (x_i - M_X) \cdot (y_i - M_Y)}{\sqrt{n \cdot \sigma_X \cdot \sigma_Y}}$$

This is an equivalently modified formula from [Lagutin, 2012]. In our further tables, this variant of the coefficient *will have the number* 1.

As we already said, in Section 3 our version of calculating the pair correlation will also be given. We note in advance that in our further tables and program fragments (Fig. 2), our variant of the coefficient *will have the number* 4.

3 On the rank correlation: our approach

In this section, we propose our own version for such a calculation; it differs significantly from Spearman's and Kendall's versions.

As input data, we obtain two different sequences of values for the same sequence of numbers. For these two sequences, we calculate the pair correlation in all the methods described above (recall that they were designated from (0) to (3)), and, in addition, we also use method (4), which we shall briefly describe further. We also remind you that in this method, we tried to take into account both the relative values of the elements in pairs (like methods (1), (2) and (3)) and their exact values (like method (0), i.e., in the case of the usual calculating the correlation coefficient).

Thus, like methods (2) and (3), we consider the set of pairs of pairs: the first pair is X_i and X_j (for random variable X implementations), and the second one is Y_i and Y_j (for Y). Similarly like methods (2) and (3), each value can be in the range from -1 to 1 (with the usual meaning of these values), and the final correlation value is obtained by averaging all obtained values. Let us also note that we are averaging the values in all pairs. Thus, when considering examples, where each sequence of consists of $3\,000$ values (such dimensions are often found in our practical tasks), there are in total $\approx 4\,500\,000$ pairs of such values for averaging.

For these pairs, we obtain the value shown on the following Fig. 4. In it, values X_i and X_j are on the left side, and values Y_i and Y_j are on the right side.

It is important that $X_i \leqslant X_j$ and $Y_i \leqslant Y_j$ (otherwise, we change *its order*, *changing* also the sign of the answer), and $X_j - X_i \leqslant Y_j - Y_i$ (otherwise, we change *the names*, *not changing* the sign of the answer). The answer is

$$R = \frac{\delta_A \cdot S}{\delta_B \cdot (S+1)}\,, \text{ where } S = \frac{\delta_A^2}{2\delta_\delta} \text{ and } \delta_\delta = \delta_B - \delta_A\,;$$

two other values are shown on the figure. This mini-algorithm is also shown in C++ on the following Fig. 2.

Let us consider some examples of our version of pair correlation for some specific pairs of value pairs. The captions to the above figures show whether we observe a strong, medium or small correlation value, including figures for degenerate cases.

Firstly, consider Fig. 5. Both examples correspond to the same order of elements in pairs (as well as all further drawings, otherwise we change the sign of the answer), but at the same time in one of the sequences, the difference in the values of the elements is much smaller than in the other. As expected, the correlation value is positive, but very small.

```
if (nReg==2) return
    (pairOne.GetA()-pairOne.GetB())*(pairTwo.GetA()-pairTwo.GetB()) < 0 ?
        -1 : 1;
    // -1 if the pair is "incorrect" and +1 if it is "correct"
else if (nReg==3) { // a more complicated version of the previous one:
    // we take into account the equality of 0 in one of the pairs
    double rOne = pairOne.GetA()-pairTwo.GetA();
    if (::IsNulla(rOne)) return 0;
    double rTwo = pairOne.GetB()-pairTwo.GetB();
    if (::IsNulla(rTwo)) return 0;
    return (rOne*rTwo) < 0 ? -1 : 1;
}
```

Figure 1. The part of the text of the function for the modificated Kendall's correlation coefficient

```
bool bOrder = true; // by default, the correct order is in both pairs
double A1 = pairOne.GetA(), B1 = pairOne.GetB(),
       A2 = pairTwo.GetA(), B2 = pairTwo.GetB();
if (A1<A2) { Swap(A1,A2); Swap(B1,B2); bOrder = !bOrder; }
if (B1<B2) { Swap(B1,B2); bOrder = !bOrder; }
// we obtained A1>=A2, B1>=B2,
// and if !bOrder then we make the negative answer
double deltaA = A1-A2, deltaB = B1-B2;
if (deltaA>deltaB) { Swap(A1,B1); Swap(A2,B2); Swap(deltaA,deltaB); }
// we obtained deltaA<=deltaB,
// but we do not change bOrder here!
if (::IsNulla(deltaA)) return (bOrder ? deltaB : -deltaB);
double deltadelta = deltaB-deltaA;
if (::IsNulla(deltadelta)) return 0.0;
double double Return = (deltaA*S)/deltaB*(S+1.0);
return (bOrder ? Return : -Return);
```

Figure 2. The part of the text of the function for the proposed calculation of the pair correlation

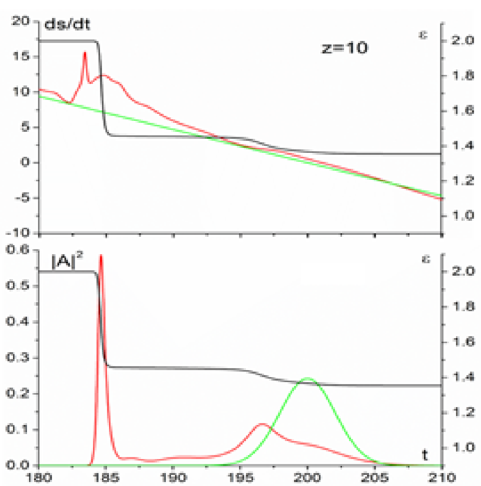

Figure 3. Frequency chirp and pulse shape

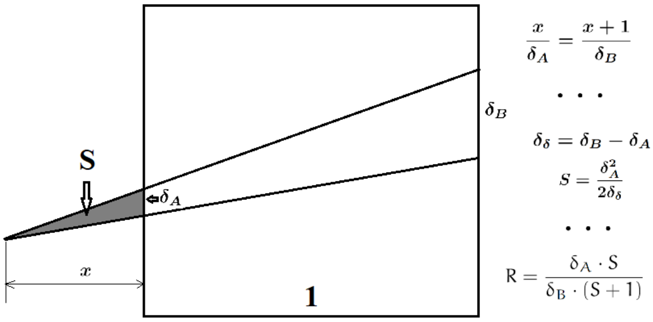

Figure 4. The proposed calculating the pair correlation

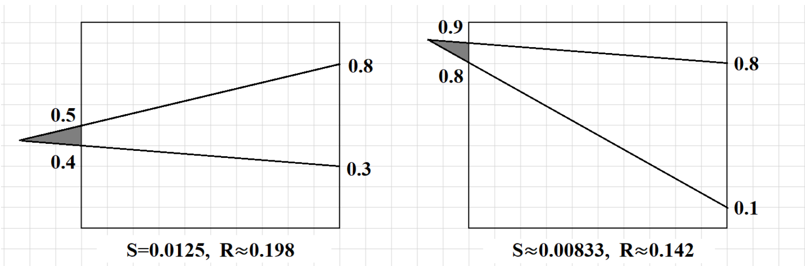

Figure 5. Examples of calculating values for the observed “small” correlation

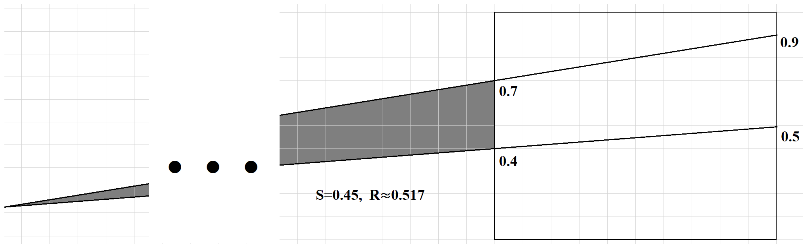

Figure 6. Example of calculating value for the observed “big” correlation

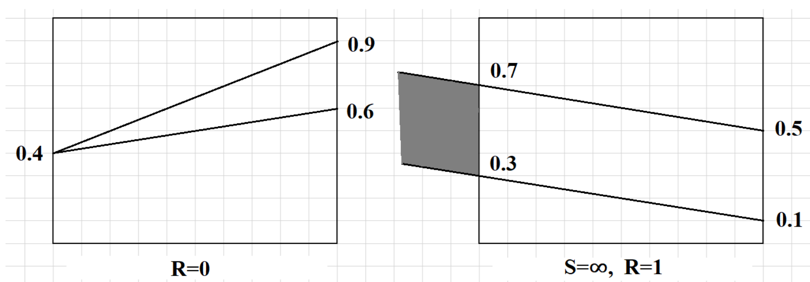

Figure 7. Example of calculating value for the degenerate cases

Secondly, consider Fig. 6. It corresponds to the case, when the difference of the same values is much more. As expected, the correlation value is more than 0.5.

Thirdly, consider two extreme cases, Fig. 7.

At the end of reviewing these examples, we note the following. In all the examples (excluding the left degenerate case, see the left part of Fig. 7), it makes sense to consider only the methods of calculating the correlation (4) and (0) (see Section 4); the other methods, i.e. (1), (2) and (3), are not meaningless, but make some sense only when considering more than one pairs of values. Thus, each time, we can use the above formulas to calculate the usual value of the correlation coefficient $R_{(0)} = 0.5$. We consider the values we receive to be closer to the truth.

Let us also remark that we do not have to count it: we understand from the statistics course that each time this value turns out to be equal $R_{(0)} = 0.5$, excluding the left degenerate case only.

4 The point of view of nonlinear optics

In this section, we describe our task in more detail than in Introduction from the point of view of nonlinear optics; at the same time, we pose the problem of why the observed and predicted results should be investigated using correlation analysis.

As we already said, within the framework of a slowly varying envelope, the process of femtosecond laser pulse propagation in the medium with golden nanorods can be described by a nonlinear Schrödinger equation with respect to a slowly varying amplitude $A(t, z)$ of laser radiation supplemented with an ordinary differential equation for nanorods aspect ratio $\varepsilon(t, z)$, [Kim et al., 2008; Driben et al., 2009; Zijlstra et al., 2009] etc. The numerical solution of these equations allows one to obtain the shape of the pulse (i.e., intensity distribution $|A(t, z)|^2$ along the time coordinate t) and the pulse phase $s(t, z)$ or frequency chirp (instantaneous frequency $\frac{\partial s(t,z)}{\partial t}$) distribution along the time coordinate at each section $z =$ const of the medium. A nonlinear frequency chirp characterizes propagation of laser radiation in a nonlinear medium (i.e., with golden nanorods). A linear frequency chirp characterizes a linear medium (without nanorods). Thus, the degree of chirp nonlinearity characterizes the degree of interaction of laser radiation with the medium.

As an example, we used the data form papers [Driben et al., 2009; Park et al., 2020]. Fig. 3 shows

- frequency chirp $\frac{\partial s(t,z)}{\partial t}$ (upper picture)
- and pulse shape $|A(t, z)|^2$ (bottom picture)

for femtosecond laser radiation propagation

- in a medium with golden nanorods (red lines)
- and in a linear medium (green lines).

Nanorods aspect ratio distribution $\varepsilon(t, z)$ is shown in both pictures by black lines and it reveals the two areas of rapid change which correspond to the laser subpulses interaction with nanorods. The largest divergence from the linear chip takes place in these areas of aspect ratio rapid change.

Simplifying the situation with Fig. 3 somewhat, we can say that there is a dependence with a detuning of the green graph on the red one. Therefore, when considering this figure and such comments, the following assumption arose. As we already said, it is possible to measure the degree of nonlinearity by calculating a correlation coefficient between the chirp distribution in a linear and nonlinear medium. Then we shall consider the correlation between the red and green lines *in different ways*,

We can say that we have actually obtained statistical characteristics similar to those assumed before the calculations. At the same time, the most adequate variant of the pair correlation coefficient is obtained in the case of the algorithm proposed by us for calculating it.

5 Most important results of computing experiments

In this section, we give some results of computing experiments for the real data previously given in Section 4. As we said before, this section is the main one of the paper, despite its small volume.

We consider the pairs corresponding to the above graphs, exactly, the values of the “green” and “red” ordinates corresponding to the abscissas from 180 to 210 of Fig. 3. We take 2460 values with the corresponding step, it turns out to be a little more than 0.0122. Of course, it is impossible to cite all values in the text of the paper [1], we shall only present the results of calculations of correlation coefficients:

```
corr-0: 0.975977
corr-1: 0.982536
corr-2: 0.938336
corr-3: 0.938335
corr-4: 0.733482
```

Let us note, among other things, the almost complete coincidence of the `corr-2` and `corr-3` variants (which is natural for real values, but does not clarify the example under consideration), as well as the expected much less change of the `corr-4` variant after normalization (0.731 instead of 0.733). Anyway, we see this variant `corr-4` as the most appropriate: *it reveals the difference between the two frequency chirp distributions*, thus differentiating the propagation in a linear and nonlinear media with sufficient accuracy.

Indeed, the difference between the laser pulse propagation in linear and nonlinear medium is significant, as it is well seen in the Fig. 3. It is very important that two subpulses, i.e., fast and slow ones, occur in the nonlinear medium, corresponding to the areas of nanorods melting. The existence of two subspulses is well marked by the chirp nonlinear distribution and its detuning from the

[1] The authors can send us the corresponding file if requested by e-mail. At the same time, almost all the data can be found at the following link: `https://owncloud.nano.sfedu.ru/index.php/s/MmTni2XsDwiYB2z`.

linear chirp distribution in the linear medium. In the fast subpulse area, the difference between the linear and the nonlinear media is most pronounced. In this area, the chirp distribution in the medium with nanorods is non-monotonic and is up to two times higher than the chirp in the linear medium. Obviously, the pair correlation between the chirps, which is equal to unity, cannot reveal the difference in the chirp distributions. *Consequently, it cannot differentiate adequately the linear and the nonlinear media.*

6 Conclusion

Let us formulate some direction for the future work.

Not very complicated, but rather long direction of work we consider the such one. We have to compare two sub-variants of our calculation of pairwise correlation given in the paper. The results differ quite strongly, but we have given only one of these sub-variants in the paper, and for the second one we have specified the formula only, see Section 3 for both the sub-variants.

Even more important for future work, we consider the exact formulation of a set of requirements to describe the computed value for only one pair of element pairs like (1). Anyway, after that we we are going to calculate the sum of these "simple" coefficients and divide the result by the value $\frac{N\cdot(N-1)}{2}$, also like Section 3. Indeed, the formulas given in [Lagutin, 2012, p. 346], passed off as *a universal variant* of counting of the pair correlation, in fact, of course, are not universal, because of the following. For instance, we want to make a function depending on *two* differences, i.e. let $c_{ij} = X_i - X_j$ (notation used by [Lagutin, 2012]); then we *cannot make an arbitrary such function* here, but must be guided exclusively by the proposed formula.

Also note that the connection of the variants of paired correlation algorithms considered in the article with discrete optimization problems was considered, for example, in [Melnikov, 2006].

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