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Multidimensional system of equations with XY-Y differentiation of probability densities p0-p1 for identification of gold nanoparticles

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ABSTRACT

The sensitivity of identification of biologically active cancer cells of metal gold nanoparticles with simultaneous differentiation by X-Y in one equation and the derivation of the second equation of Y-density of probability p0-p1 of intersection of ellipses of Raman scattering of statistical values of peaks of spectra for formulation and solution of a multidimensional system of equations are estimated. For the probability density p0-p1 in the usual system of nonlinear equations is the intersection of the ellipses of the distribution of the accuracy of the solution $(-2.878 \div -9.989)\cdot 10^{-4}$ (r0=0.038208 and R1=0.038235). The accuracy of the solution of systems of differential equations of the ellipses of the distributions of XY and Y increases to $(-5.20\cdot 10^{-18}$ $-7.59\cdot 10^{-19})$. In this case, there is uncertainty in determining the density of the probability of intersection of ellipses and identified 2 intersection points with solutions p0=0.00849717886507494 and p1=0.13505614093891083. When checking solutions for graphical evaluation of intersection of the ellipses for the distribution of the spectra of Raman scattering was discovered a significant error at 87.2% when identifying p0 and p1. A numerical additive Δ=0.02152711027273521 was introduced into the differential equation for Y to eliminate uncertainty. When solving this system of differential equations with additive accuracy $(6.94\cdot 10^{-18} \div -1.22\cdot 10^{-18})$ the probability density is determined with sufficient accuracy $5.447366606424859\cdot 10^{-16}$: p0=0.03821421104129397 and p1=0.03821421104129395

Keywords: *gold nanoparticles, multi-dimensional differential system via correlation equations XY-Y polyester fiber, polarization spectra of Raman scattering, the accuracy of detection, the probability of intersection of the ellipses of the distribution, the accuracy of the identification.*

1. INTRODUCTION

To ensure the resolution of 10^{-9} - nanomolar and 10^{-12} - picomolar ability in the analysis of biological objects, it is necessary to use Raman spectroscopy with surface enhancement on gold nanoparticles [1, 2], as well as the use of mathematical processing of experimental data by multivariate vector - matrix correlation [3, 4]. Multidimensional vector-matrix correlation methods of data processing increase the reliability of identification up to 10^{-16} [4, 5].

Raman spectroscopy with surface electromagnetic plasma amplification (SERS) is increasingly used in fundamental physics and chemistry, as well as in materials science, environmental sciences, biology, biophysics and medicine [6-18]. SERS can be used as a disease detection method capable of providing ultra-high sensitivity in biomolecular and chemical sensing [19]. The plasmon effect of SERS is detected on noble metal nanoparticles (Au, Ag, Pt, etc.), and the shape of these nanoparticles can be very diverse: from the complex shape of surfaces to aggregates of nanoparticles [20-23].

To date, a large number of studies have been conducted on the high reliability and high resolution of nanostructured SERS nanoparticles, which can provide electromagnetic amplification with surface plasmon resonances and chemical amplification due to charge transfer processes. In addition to noble metals, graphene, semiconductors and transition metal oxides are beginning to be used for SERS as potential SERS-active substrates [16, 19, 20]. The combination of electromagnetic and chemical effects can make SERS a powerful method to detect vibrational spectra even from individual molecules and identify them in microfluidic devices of biology, chemistry and medicine.

In recent years, the SERS method has been widely used in the medical field as a powerful spectroscopy as a diagnostic tool for diseases, as well as the detection and monitoring of nanopreparations in the treatment of cancer [6, 14-16].

For pre-testing of mathematical models identification of gold nanoparticles with the use of Raman spectroscopy uses the dielectric fiber: acrylic, dacron, polyester, cotton, etc. Most acceptable to identify the gold nanoparticles in the simplicity of the Raman spectrograms is polyester fiber. The most complex spectrogram has cotton.

Estimation of reliability of detection of gold nanoparticles on textile materials is carried out in [3, 4, 24, 25]. Since the presence of nano-gold slightly changes the intensity of the Raman spectra of polyester fiber in comparison with nanosilver, it is necessary to develop a new method for the formulation and solution of a system of multidimensional equations. Since the manual selection of the probability density of the intersection of the distribution ellipses is highly complex, the solution of the problem is extremely inconvenient and quite slow in time.

For Fig. 1 and Fig. 3 presents the results of validation for the solution of the problem of sensitivity with the detection of densities of probabilities of the intersections of the ellipses of the distribution. Using eigenvalues and fundamental matrices, the method of generation of multivariate statistical correlation data is applied. Generation held for 9 peaks of the spectrum of Raman scattering light at the same time for fibers without and with gold nanoparticles [25]. The intersection of the distribution ellipses was estimated in a large range of the number of generated data up to 10 million. In this case, the intersection ellipses for any nonlinear transformation will be described by the normal multidimensional distribution law. This greatly simplifies the use of fundamental correlation matrices. But the use of this method is not enough for reliable detection of gold nanoparticles, since the accuracy of identification is 10^{-4}-10^{-5}.

In [3, 24, 25] the methods of preparation and analytical solution of a system of multidimensional nonlinear equations for the identification of gold nanoparticles are considered . The accuracy of estimating the probability density at the point of intersection are identical, and the accuracy of the solution is equal to $(-2.878 \div -9.989)\cdot 10^{-4}$. To solve a system of nonlinear equations requires a sufficiently long manual selection of parameters. Therefore, such accuracy is not enough to detect gold nanoparticles.

The paper assesses the applicability of the equations to solve the system in the differentiation of the XY equation in the bundle, as well as in the differentiation of the second equations in y unknown analytical ellipses of probability densities p0-p1 of the intensity distribution of the polarization spectrum peaks of Raman scattering in the identification of gold nanoparticles on the surface of polyester fibers.

In this paper, in order to further use the system of quadratic differential correlation equations in the system of equations with a large number of unknowns up to 9, we propose a method of compiling a system of equations with only two unknowns when solving the problem in vector-matrix form.

2. EXPERIMENTAL SECTION

Measurement of statistical data on the intensities of the spectra of polyester fibers with and without nanoparticles revealed the distribution parameters and correlation matrices of all 9 major peaks taking into account the polarization of radiation by X and Y (1) and Fig.1-2.

In [24] confirmed that the spectrograms are significantly different in the intensity of peaks of the spectra for polarization of the Raman radiation X - across the grain-and Y - along the fibers. In the Y direction, the peaks have intensity 2-3 times greater than in the X direction for both fibers without nanoparticles and fibers with nanoparticles. This is explained by the fact that the length of the Raman irradiation zone along the fibers is much longer than across the fibers.

In this paper, for gold nanoparticles due to the sufficient complexity of measurements on the Raman spectrometer with a repetition of 5-10 times for different zones of the fiber surface, the method of statistical data generation on mathematical expectations, mean square deviations (1) and correlation matrices (Fig. 2).

This stage of work is necessary for a graphical preliminary assessment of the intersection of the ellipses of the intensity distribution of the Raman spectra peaks of polyester fibers with and without gold nanoparticles.

The method of statistical data generation using double transformation of fundamental matrices was developed in [24]. It is applied to improve the accuracy simultaneously for 9 peaks in X and Y directions of polyester fiber with nanoparticles and without gold nanoparticles. The number of generated data can vary from 12 to 10 million.

Figure 1 shows 48 generated Raman scattering statistics: - polyester fiber with gold nanoparticles $Tr^{(n)}$ at polarization across X and $TrY^{(n)}$ along Y fibers; - polyester fiber without gold nanoparticles $Dr^{(m)}$ at polarization across X and $DrY^{(m)}$ along Y fibers; - n and m numbers of spectral peaks.

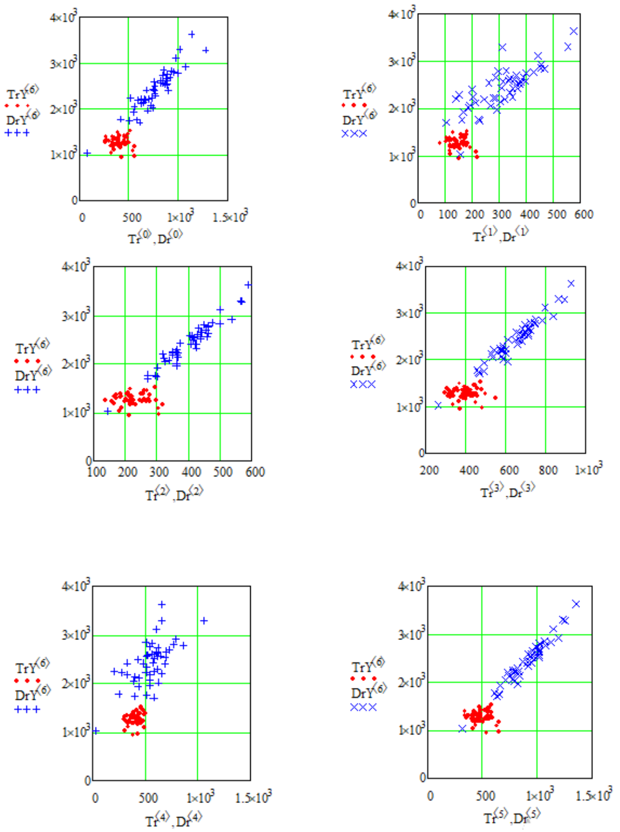


Figure 1. Statistical spectral data of Raman scattering: - polyester fiber with gold nanoparticles Tr $^{(n)}$ at polarization across X and $TrY^{(n)}$ along Y fibers.
- polyester fiber without gold nanoparticles Dr $^{(m)}$ at polarization across X and $DrY^{(m)}$ along Y fibers; - n and m peak numbers.

Figure 3 shows 48 and 96 generated Raman scattering statistics and the tangents at the intersection of the distribution ellipses with gold nanoparticles and without nanoparticles.

In Fig.1 and Fig. 3 it can be seen that in the one-dimensional case by projection on the X-axis ($Tr^{(n)}$ and $Dr^{(m)}$) the data will completely intersect with a probability of about 1 and the information content of the intersection is very low.

The data will intersect on the projection on the Y-axis (TrY(n) and Dre (m)) with probability 1/48=0.0208, and the information content of the intersection is very large.

When using a two-dimensional method of intersection of ellipses is taken into account the simultaneous intersection of the X-axis, Y-axis and the correlation between the data, therefore, the distribution of information will be even higher.

Hence, there is a need to apply multidimensional ellipsoid equations of order 9, taking into account the multidimensional correlation.

Figure 3a shows that data intersect only in a single case for 48 data, and for 96 data in figure 3b occurs in several cases.

$$rXY1 := \begin{pmatrix} -0.037942 & -0.059351 & -0.036499 & -0.065879 & -0.15344 & -0.01173 & -0.143937 & 0.102304 & -0.056958 \\ -0.124291 & -0.14975 & -0.11752 & -0.153551 & -0.242178 & -0.100427 & -0.230521 & 0.027481 & -0.051446 \\ 0.040257 & 0.021687 & 0.039271 & 0.012677 & -0.078362 & 0.069321 & -0.072415 & 0.178587 & -0.060715 \\ 0.005592 & 0.000983 & -0.008381 & -0.016315 & -0.096501 & 0.041525 & -0.098049 & 0.12117 & -0.283381 \\ 0.403357 & 0.386308 & 0.388069 & 0.393493 & 0.387607 & 0.397642 & 0.418896 & 0.372158 & 0.482458 \\ -0.112396 & -0.131075 & -0.114448 & -0.138566 & -0.218647 & -0.086114 & -0.208227 & 0.019404 & -0.132342 \\ 0.133612 & 0.098623 & 0.144516 & 0.110096 & 0.058397 & 0.132967 & 0.087183 & 0.211087 & 0.436028 \\ 0.084858 & 0.045002 & 0.103128 & 0.068248 & 0.052881 & 0.06455 & 0.091762 & 0.113937 & 0.634366 \\ -0.006686 & -0.047192 & 0.013172 & -0.023994 & -0.040852 & -0.026223 & -0.00194 & 0.032552 & 0.574372 \end{pmatrix}$$

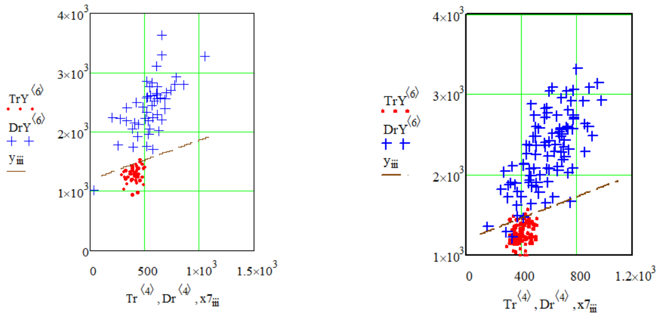
a

$$rXY0 := \begin{pmatrix} 0.919749 & 0.910703 & 0.93322 & 0.91605 & 0.930313 & 0.903024 & 0.934089 & 0.910258 & 0.901971 \\ 0.898723 & 0.918213 & 0.894694 & 0.908353 & 0.896547 & 0.911643 & 0.888256 & 0.874308 & 0.661293 \\ 0.990871 & 0.993788 & 0.993876 & 0.990894 & 0.994549 & 0.986498 & 0.992762 & 0.955976 & 0.89505 \\ 0.984181 & 0.987972 & 0.99193 & 0.985103 & 0.991022 & 0.977624 & 0.989289 & 0.961217 & 0.904798 \\ 0.659208 & 0.635798 & 0.675349 & 0.646855 & 0.672121 & 0.63041 & 0.681502 & 0.644343 & 0.796864 \\ 0.991028 & 0.992417 & 0.997576 & 0.992284 & 0.996638 & 0.985381 & 0.995919 & 0.972923 & 0.87666 \\ 0.577047 & 0.54968 & 0.594581 & 0.563624 & 0.590666 & 0.545919 & 0.601809 & 0.569881 & 0.731272 \\ 0.460466 & 0.431792 & 0.47361 & 0.443309 & 0.471808 & 0.428492 & 0.482841 & 0.430613 & 0.683283 \\ 0.454615 & 0.425248 & 0.475784 & 0.440676 & 0.470541 & 0.420535 & 0.482834 & 0.459426 & 0.646792 \end{pmatrix}$$

b

Figure 2. Correlation matrices of Raman polarization spectrograms of polyester fibers after drying in natural conditions in the frequency range 54.3÷3110см$^{-1}$: a - polarization across and along the fibers with gold nanoparticles; b) polarization across and along fibers without gold nanoparticles.

The solution of the system of equations presented in Fig. 4 and 5, tangent b at the intersection point shows that the derivatives will be equal for ellipses with both nanoparticles and non-nanoparticles.



a b

Figure 3. Statistical spectral data of Raman scattering and tangents at the intersection of distribution ellipses with gold nanoparticles and without nanoparticles: a-48 data; b-96 data.

There is a need to compile a system of equations obtained in the form of X and Y. This is a difficult task, but it makes it possible to improve the multidimensionality of the system of equations.

In order to significantly reduce the complexity of the preparation and solution of a system of nonlinear equations, it is necessary to apply the simultaneity of XY derivatives in the same system in the same equation of the derivative Y.

In Fig. 1, 3 and 5 defined derivative for the tangent equation at the intersection of statistical data y=0.675·(x-411.35)+ 1458.69.

The accuracy of determining the derivatives in the analytical determination is 10^{-15}. In the analytical determination of derivatives in the equations of tangents for fibers without nanoparticles and with nanoparticles, they will have the following form:

y0=0.6764577648523631 ·(x0-411.3526470394268) + 1458.6903699336267;
y1=0.6764577648523626 ·(x1-411.3526470394268) + 1458.6903699336267.

$$\begin{aligned} MENX^T &= (416.914 \quad 152.378 \quad 227.443 \quad 400.725 \quad 398.947 \quad 502.157 \quad 514.102 \quad 278.196 \quad 499.456); \\ MENY^T &= (525.478 \quad 321.605 \quad 854.972 \quad 2861.032 \quad 2872.996 \quad 1258.934 \quad 1268.02 \quad 143.918 \quad 151.002); \\ \sigma\Delta X^T &= (70.798 \quad 27.978 \quad 41.622 \quad 53.385 \quad 47.117 \quad 72.298 \quad 67.912 \quad 35.079 \quad 56.89); \\ \sigma\Delta Y^T &= (50.752 \quad 29.74 \quad 79.231 \quad 259.348 \quad 270.86 \quad 100.309 \quad 113.697 \quad 6.59 \quad 14.109); \\ MENX0^T &= (745.336 \quad 307.767 \quad 387.809 \quad 626.062 \quad 569.689 \quad 868.354 \quad 771.902 \quad 405.376 \quad 748.634); \\ MENY0^T &= (972.254 \quad 606.828 \quad 1598.202 \quad 5178.102 \quad 5128.383 \quad 2304.268 \quad 2288.106 \quad 238.792 \quad 234.17); \\ \sigma\Delta X0^T &= (196.404 \quad 93.691 \quad 81.964 \quad 121.371 \quad 166.386 \quad 186.764 \quad 241.501 \quad 89.597 \quad 175.22); \\ \sigma\Delta Y0^T &= (221.73 \quad 133.433 \quad 358.411 \quad 1038.482 \quad 1030.864 \quad 473.795 \quad 455.166 \quad 35.978 \quad 32.436). \end{aligned} \qquad (1)$$

3. RESULTS SECTION

Preparation and solution of quadratic differential equations of the correlation of the random values of the distribution peaks of the Raman spectra of Raman radiation was conducted on the preliminary experiments with the use of correlation matrices and the parameters of the distributions taking into account the polarization of X - across and Y - along the fibers simultaneously in one measurement [24, 25].

It was also found that when solving a system of differential correlation equations by other methods, uncertainty was obtained, consisting in the fact that when checking the solution for the graphical intersection of distribution ellipses without nanoparticles and with nanoparticles, two intersection points appeared. The values of the ellipse intersection probability densities were not accurate enough [3-4]. Such a solution for the identification of gold nanoparticles is unacceptable and can be used only for preliminary results.

When compiling a system of equations in vector-matrix correlation analytical expressions by the coordinates of the intersection points with differentiation by XY and Y, the p-density of the probability of intersection of the distribution ellipses is used. In this study, we work with a system of only two vector-matrix analytical expressions of probability densities $p0=g(X0^T\cdot\sum0^{-1}\cdot X0)$ and $p1=f(X^T\cdot\sum1^{-1}\cdot X)$ [14-16].

Differential correlation equations f (x,y) and g(x,y) [4-5] are obtained by differentiating the original equation F (x,y) by Y and differentiating simultaneously by X and Y . In the equation we need to introduce the additive Δ. As a result of the solution of the proposed system of equations, the accuracy is determined: f(v0, v1)= 6.94·10^{-18} and g(v0, v1)= -1.22·10 $^{-18}$ (3). It shows obtaining precision p0 and p1 to 7.263155475233148·10^{-16}. The accuracy for estimating the intersection coordinates of the distribution ellipses i=4 and j=6 is also high:

$$\sum 0 := \begin{pmatrix} 1 & rXY0_{i,j} \\ rXY0_{i,j} & 1 \end{pmatrix} \qquad \sum 1 := \begin{pmatrix} 1 & rXY1_{i,j} \\ rXY1_{i,j} & 1 \end{pmatrix}$$

$$X0 := \begin{pmatrix} \dfrac{x - MENX0_i}{\sigma\Delta X0_i} \\ \dfrac{y - MENY0_j}{\sigma\Delta Y0_j} \end{pmatrix} \qquad X := \begin{pmatrix} \dfrac{x - MENX_i}{\sigma\Delta X_i} \\ \dfrac{y - MENY_j}{\sigma\Delta Y_j} \end{pmatrix} \qquad (2)$$

$$f(x,y) := \left[\frac{d}{dy}\left[\ln\left[\frac{\left[\left(\left|\sum 1\right|\right)^{-0.5}\right]}{(2\cdot\pi)}\right] + \frac{-1}{2}\cdot\left(X^T\cdot{\sum 1}^{-1}\cdot X\right)\right]\right]$$

$$-\left[\frac{d}{dy}\left[\ln\left[\frac{\left[\left(\left|\sum 0\right|\right)^{-0.5}\right]}{(2\cdot\pi)}\right] + \frac{-1}{2}\cdot\left(X0^T\cdot{\sum 0}^{-1}\cdot X0\right)\right]\right] + 0.0215271102\ 7273520976$$

$$g(x,y) := \begin{bmatrix} \dfrac{d}{dx}\left[\dfrac{-1}{2}\cdot\left(X^T\cdot{\sum 1}^{-1}\cdot X\right)\right] & \dfrac{d}{dy}\left[\dfrac{-1}{2}\cdot\left(X^T\cdot{\sum 1}^{-1}\cdot X\right)\right] \\ \dfrac{d}{dx}\left[\dfrac{-1}{2}\cdot\left(X0^T\cdot{\sum 0}^{-1}\cdot X0\right)\right] & \dfrac{d}{dy}\left[\dfrac{-1}{2}\cdot\left(X0^T\cdot{\sum 0}^{-1}\cdot X0\right)\right] \end{bmatrix} \qquad (3)$$

x:= 390.0 y:= 1500.0

Given

f(x,y) = 0 g(x,y)= 0

v2:= Find(x,y)

$$v2 = \begin{pmatrix} 411.3526470394\ 268 \\ 1458.6903699336\ 267 \end{pmatrix}$$

$f(v2_0,v2_1) = 6.9388939039072275\cdot10^{-18}$

$g(v2_0,v2_1) = -1.2197274440461925\cdot10^{-18}$

The probability density for the intersection point of the ellipses of the intensity distribution of the Raman spectra was determined by solving equations (4-5):

$$p0 := \frac{1}{(2\cdot\pi)\cdot\left[\left(\left|\sum 0\right|\right)^{0.5}\right]}\cdot e^{\frac{-1}{2}\cdot\left[\left(\frac{v2_0-MENX0_i}{\sigma\Delta X0_i}\ \ \frac{v2_1-MENY0_j}{\sigma\Delta Y0_j}\right)\cdot\left(\sum 0\right)^{-1}\cdot\begin{pmatrix}\frac{v2_0-MENX0_i}{\sigma\Delta X0_i}\\ \frac{v2_1-MENY0_j}{\sigma\Delta Y0_j}\end{pmatrix}\right]} \qquad (4)$$

$$p1 := \frac{1}{(2\cdot\pi)\cdot\left[\left(\left|\sum 1\right|\right)^{0.5}\right]}\cdot e^{\frac{-1}{2}\cdot\left[\left(\frac{v2_0-MENX_i}{\sigma\Delta X_i}\ \ \frac{v2_1-MENY_j}{\sigma\Delta Y_j}\right)\cdot\left(\sum 1\right)^{-1}\cdot\begin{pmatrix}\frac{v2_0-MENX_i}{\sigma\Delta X_i}\\ \frac{v2_1-MENY_j}{\sigma\Delta Y_j}\end{pmatrix}\right]} \qquad (5)$$

p0 = 0.03821421104129397

p1 = 0.03821421104129395.

For the usual system of intensity equations of the Raman spectrum peaks i=4 and j=6, the probability density will have the following values: p0=0.03768476300359416 and p1= 0.03771243643792218. When the system of equations with the differentiation with respect to X to get the peaks of the Raman spectrum of i=4 and j=6 probability density p0=0.03821413373638296 and p1=0.03821413373638301. With a system of equations with differentiation by Y, we obtain for the peaks of the Raman spectrum i=4 and j=6 the probability density p0=0.03777989496468339 and p1= 0.03777974849915652.

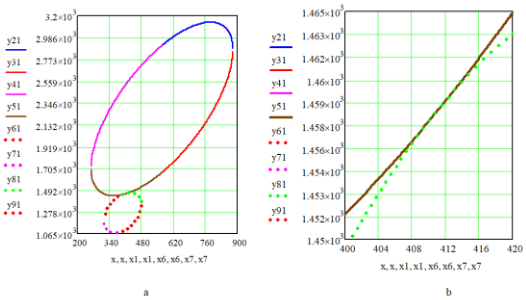


Figure 4. Graphical evaluation of the intersection of the ellipses of the Raman spectra distribution in the identification of nanoparticles in polarization Y-along and X-across fibers with differentiation by XY-Y with the result of solving the system of equations p0=0.03821421104129397 and p1=0.03821421104129395: a-general view, b-enlarged fragment of the area of intersection of distribution ellipses.

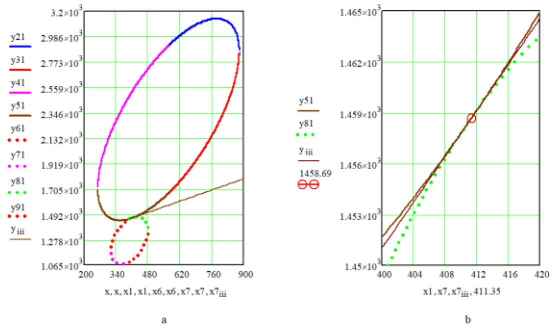


Figure 5. The intersection of the ellipses of the density probability distribution of the intensities of the Raman spectra c of the tangent at the point of intersection: a-general view; b - enlarged fragment of the region of intersection of the ellipses of the distribution of the coordinates of the intersection X=411.3526470394268, Y=1458.6903699336267.

4. CONCLUSIONS

In assessing the numerical solution of differential equations (1-5) revealed high accuracy in the results in a large range of initial parameters x=100 ÷ 412 and y=1470 ÷ 5500 with stable parameters of the solution results.

The obtained values of the coordinates of the intersection of the distribution ellipses X=411.3526470394268, Y=1458.6903699336267 is highly accurate and does not depend on the initial values of the parameters X and Y in the specified range.

The practically required accuracy of the results depends on the sensitivity of the developed method of identification of gold nanoparticles. In order to ensure the resolution of 1-2% of the concentration of gold nanoparticles on the fibers, it is necessary to process the results with an accuracy of $(1\text{-}2)\cdot10^{-4}$, and the accuracy of obtaining the value of each parameter $(1\text{-}2)\cdot10^{-6}$. For reliable detection of a nanoparticle, the accuracy of solving the system of equations 10^{-12} - 10^{-16} is required.

In this paper, the accuracy of the solution (3) f(v0, v1) = $6.9388939039072275\cdot10^{-18}$ and g(v0, v1) = $-1.2197274440461925\cdot10^{-18}$ for the initial given parameters x=390, y=1500 is revealed. At initial set parameters x=412, y=1470 the accuracy of the solution is: f(v0, v1)= $6.9388939039072275\cdot10^{-18}$ and g (v0, v1)= $9.75781955236954\cdot10^{-19}$.

The accuracy of the solution for the system of nonlinear equations for the probability densities p0 and p1 of the intersection of the distribution ellipses is $(-2.878 \div -9.989)\cdot10^{-4}$ (p0=0.038208 and p1=0.038235). When using systems of differential equations XY-Y ellipses of distributions, the accuracy of the solution is

increased to ($-5.20 \cdot 10^{-18} \div -7.59 \cdot 10^{-19}$). In this case, there is uncertainty in determining the radius of intersection of ellipses and finding solutions found 2 intersection points p0=0.00849717886507494 and
p1= 0.13505614093891083. A sufficiently large error of 87.2% of the determination of p0 and p1 was revealed when checking the solution for the graphical evaluation of the intersection of ellipses of the distribution of these Raman spectra. A numerical additive Δ=0.021527110273520976 was introduced into the differential equation for Y to eliminate uncertainty. The accuracy of the solution of this system of differential equations is ($-1.2197274440461925 \cdot 10^{-18} \div 6.9388939039072275 \cdot 10^{-18}$) with the probability density of p0=0.03821421104129397 and p1=0.03821421104129395 with their accuracy $5.447366606424859 \cdot 10^{-16}$.

The objectives of further research are: to develop a method for solving the system of equations for differentiation jointly along the X , Y and Z axes, with the number of unknown parameters more than two; to assess the reliability of the identification of gold nanoparticles on polyester fibers using multidimensional differentiation by X, Y and Z simultaneously with the study of two, three peaks of the spectrum.

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6. ACKNOWLEDGEMENTS

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