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Review Article

From desolvation-induced self-organization on the MALDI anchor target chip surfaces to laser-induced self-organization in MALDI techniques: Correlation-spectral analysis and complex wavelet analysis of tesiographic spots on the anchor chips

Theodor K. Orekhov*, Oleg V. Gradov

Institute of Energy Problems for Chemical Physics RAS, N. N. Semenov Federal Research Center for Chemical Physics of the Russian Academy of Sciences (FRC CP RAS), 119991 Moscow, Russia

*** Corresponding author:** Theodor K. Orekhov, theorehov@gmail.com

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ABSTRACT: This article proposes to analyze the formation and "morphogenesis" during desolvation of drops on MALDI targets and target chips using 2D correlation spectral analysis based on the two-dimensional Fourier transform and wavelet spectroscopy methods in the real and imaginary regions. The results of the correlation-spectral and wavelet analysis are shown in the illustrations in the text of the article.

KEYWORDS: desolvation; dehydration; LDI MS; MALDI; LAMMA; anchor chips

1. Introduction

It is well known, that MALDI MS frequently used for analysis of biological complex mixtures[1–15]. The applicability of Matrix Assisted Laser Desorption/Ionization methods (MALDI) in computer-assisted identification of different biochemical constituents on ahcnor chips (also known as MALDI target plates, including barcoded machine-readable ones)[16–18] with areas on which the corresponding samples for identification are pipetted is a matter of common knowledge. Pipetting of the sample onto the target plates/anchor chips can be performed manually with a glass or plastic tip (in the latter case, the probability of contamination of the sample with organic contaminants is often significantly increased) or automatically with special devices[19–29]. Literature on MALDI sampling is extremely large[30–49].

Ideally, if the goal is not just detection but specific quantitative or semi-quantitative data collection on the content of target substances in samples, the drops should be similar in volume and microrheological properties, which is not always possible with manual dosing on the plate. Identical drops are identically desolvated and crystallized however, drops applied at intervals in time, at any time after their pipetting (see **Figure 1**) have different optical and recrystallometric, desolvated and microrheological characteristics. From the standpoint of statistical data analysis and metrology, this can result in heteroscedasticity in the sample statistics associated with the difference in the size of drops and the completeness of filling the wells on the plate (MALDI target plates/anchor chips) after manual dosing. This problem is particularly evident when the types of wells geometries differ. In essence, the products of analyte dehydration/crystallization, both colloidal and macromolecular (e.g., polypeptide) in nature, from the standpoint of nonlinear physics, are self-organization products[50–61] that are formed in

the presence of the corresponding energy conditions/gradients, including laser-induced desolvated (dehydrated) self-organization. Therefore, the form of such self-organizing structures strongly depends on the medium conditions and the experimental protocol.

For both colloidal and supramolecular structures, the statements about the dependence of the structure on the preparation conditions are true. The transition from manual dosing to automatic pipettes for applying "spots" (also known as MALDI spotter), although it leads to the improvement of the reproducibility of dosing and uniformity of the plate in volume filling, does not lead to the elimination of the physical causes of desolvated heterogeneity. Therefore, when measuring native samples by direct mass spectrometry uniformity of spots is often neglected and the performance of a specific quantitative analysis is not considered, with the only purpose to identify the presence of a particular compound/chemical agent in the analyzed sample, or chemical identification of an unidentified sample, collected directly in the natural conditions, according to its MALDI mass spectra without an extremely complicated sample preparation, associated with chemical separation of the biological sample into elementary identifiable molecular components (lipids, proteins, etc.).

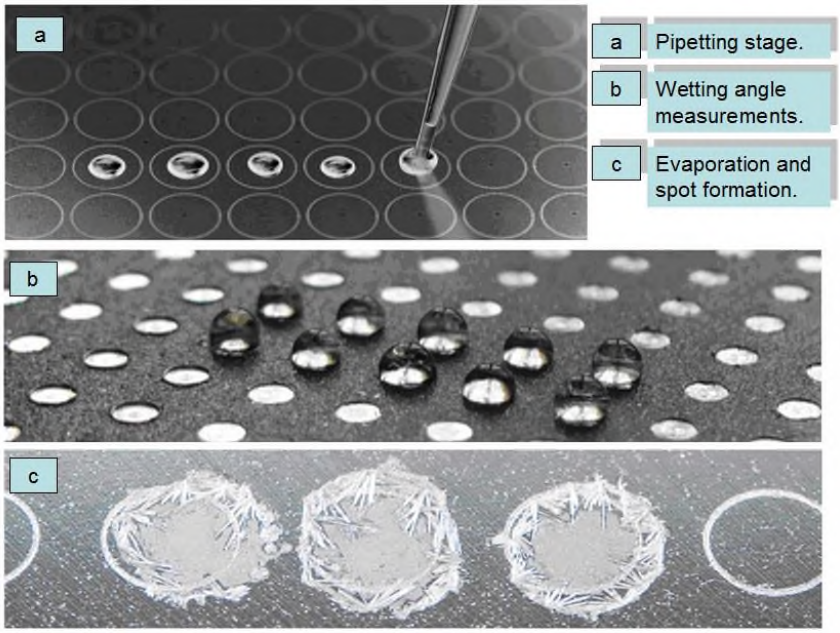

Figure 1. Stages of the liquid droplet desolvation and pattern formation in the self-organization on MALDI.

2. Methods

For a serial comparative analysis of stain dehydration patterns on MALDI chips, it was proposed to use the Fourier transform of stain images. At the same time, with the help of a neural network, each chemism of spots could be associated with a certain complex of descriptors of chemical and physical genesis. As a tool for express Fourier spectral analytics, the QAVIS software package was used, which generated IFCs and ISCs (Integral Frequency Characteristics and Integral Spatial Characteristics) based on the two-dimensional Fourier transform of the image of a spot, a drop on a chip. This complex was developed at the POI FEB RAS by the group of Fishchenko and Goncharova[62–65]. It is based on the FFTW library and was previously used for various tasks[66–72].

This technique of comparative Fourier measurements was named correlation-spectral analysis by the authors of the software package themselves.

Wavelet analysis was performed using the QAVIS program in real and imaginary coordinates.

3. Results

The results of the correlation-spectral Fourier analysis of various forms of desolvation in drops are shown in **Figures 2**–**4**. The results of wavelet correlation-spectral analysis of different forms of desolvation in drops (with separate analysis in three image fragments—on three scan strips of a photo of a drop) are shown in **Figures 5**–**7**. It can be seen that both the Fourier method and the wavelet method can distinguish between different forms of desolvation and shaping/crystallization/reaction-diffusion mophogenesis during the transition to the solid phase. This is characteristic, since when pipetting onto MALDI substrates, one can observe heteroscedasticity of samples of morphometric characteristics of droplets and patterns of their dehydration or desolvation (see **Figure 8**).

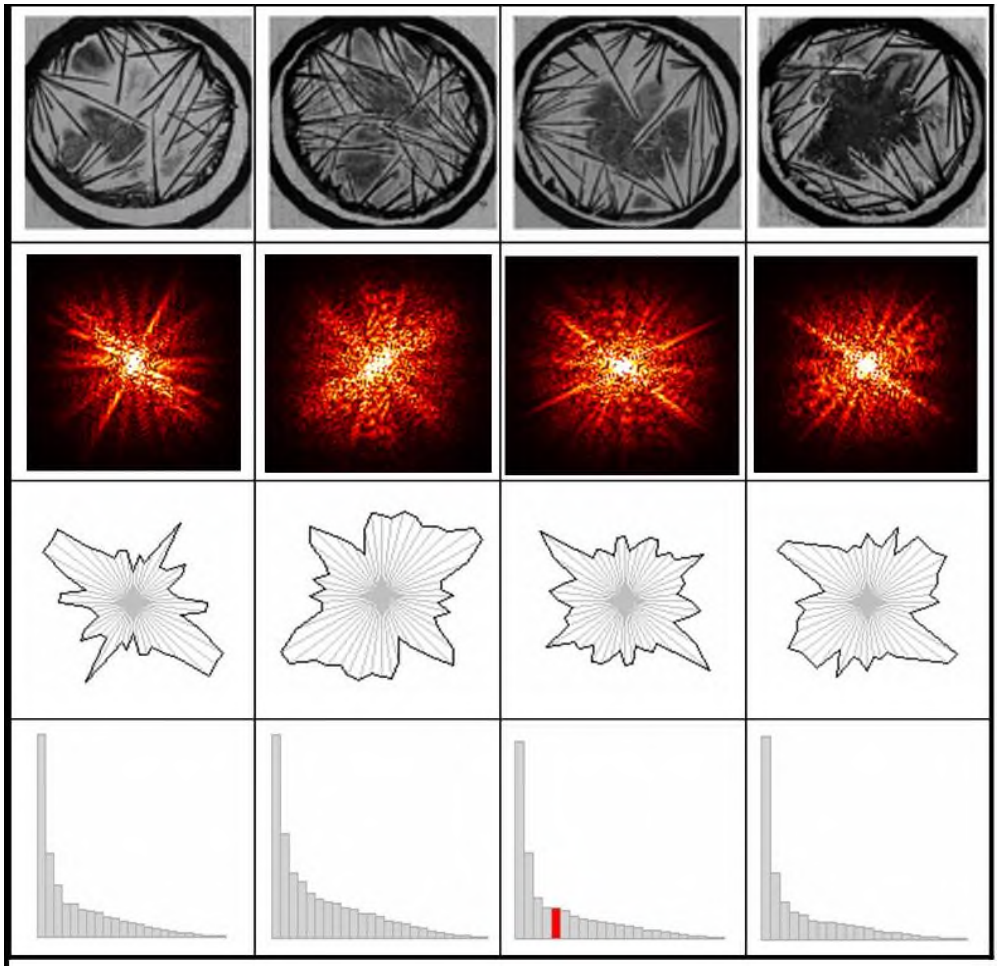

Figure 2. Comparative 2D FFT correlation-spectral analysis of droplets; example of image processing from the old presentation in Moscow State University.

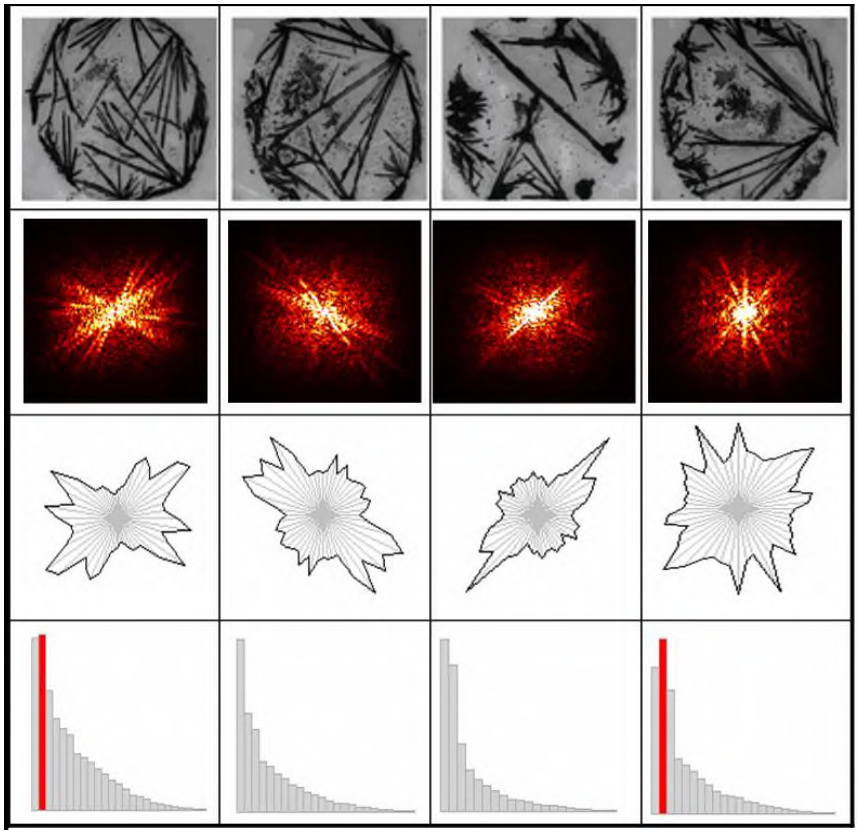

Figure 3. Comparative 2D FFT correlation-spectral analysis of droplets; example of image processing from the old presentation in Moscow State University.

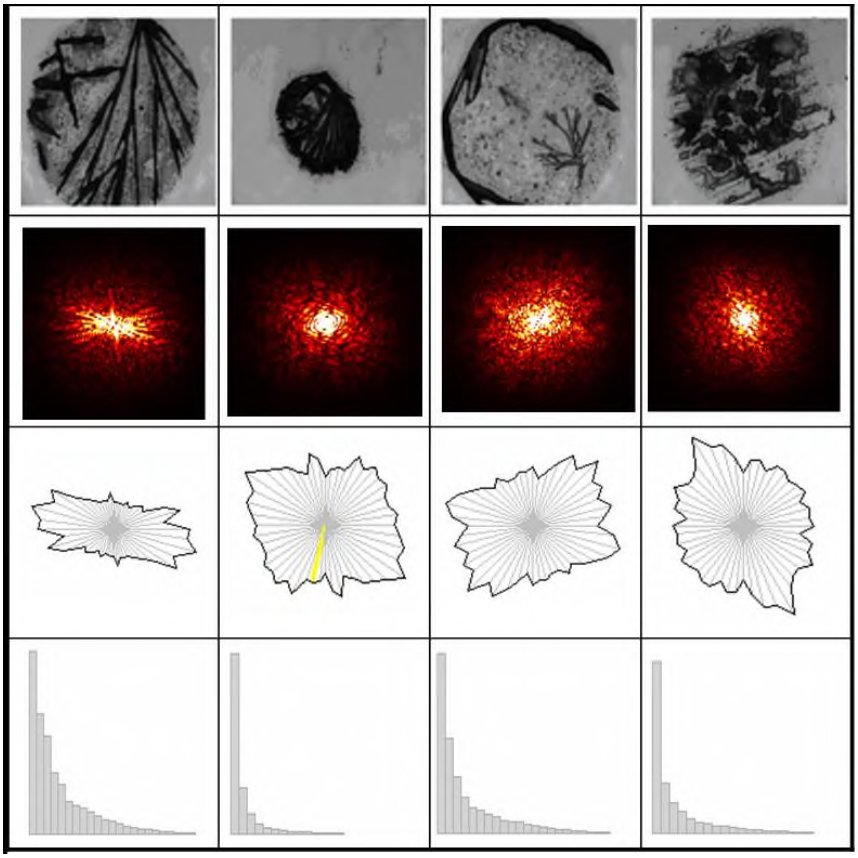

Figure 4. Comparative 2D FFT correlation-spectral analysis of droplets; example of image processing from the old presentation in Moscow State University.

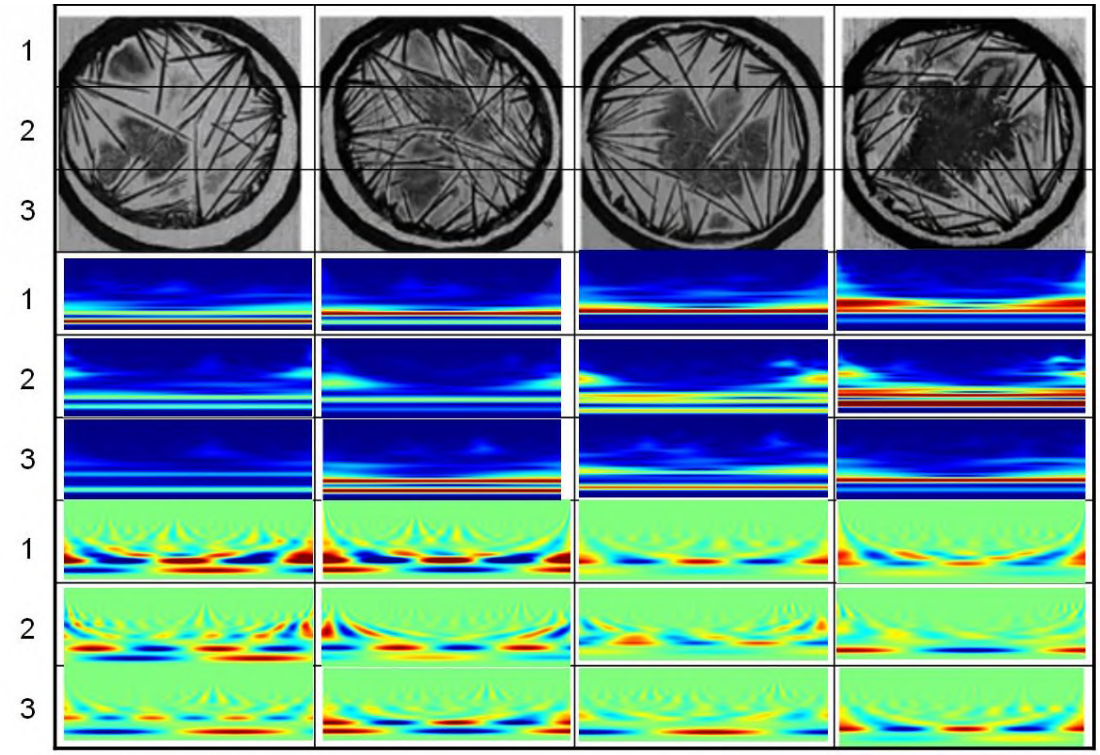

Figure 5. Comparative wavelet correlation-spectral analysis of droplet images; example of image processing from the old presentation in Moscow State University.

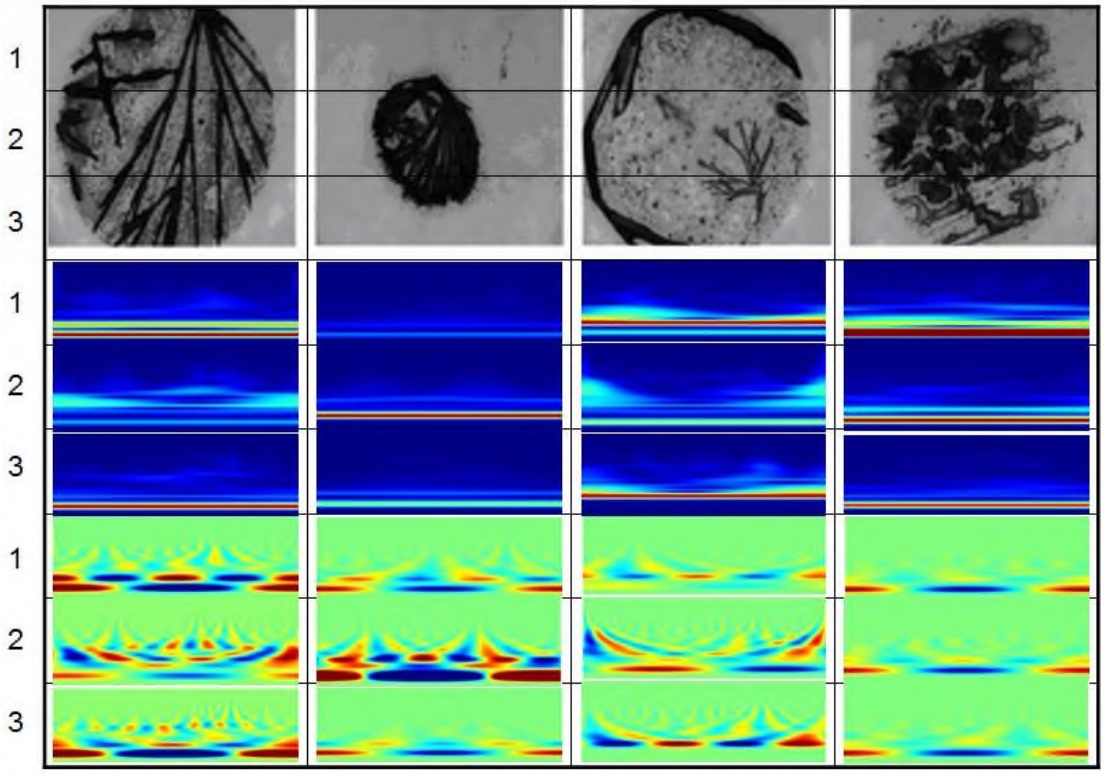

Figure 6. Comparative wavelet correlation-spectral analysis of droplet (Ibid.).

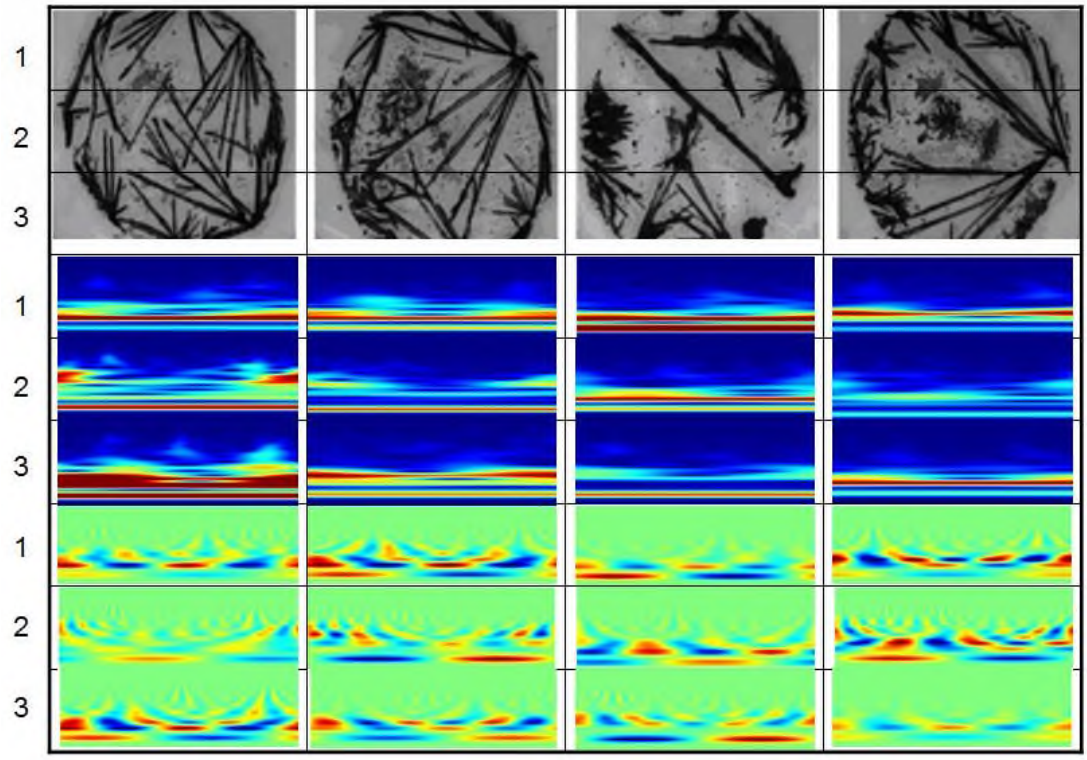

Figure 7. Comparative wavelet correlation-spectral analysis of droplet; example of image processing from the old presentation in Moscow State University.

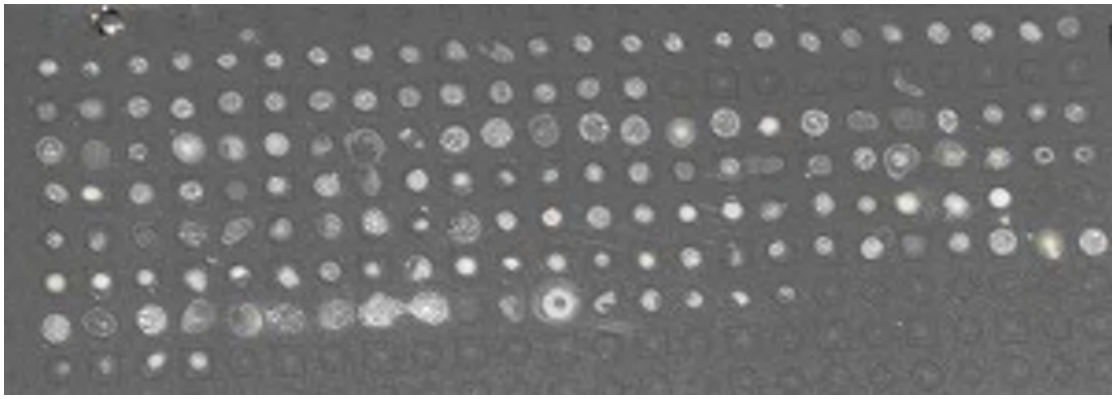

Figure 8. Comparative 2D FFT correlation-spectral analysis of droplets; example of image processing from the old presentation in Moscow State University.

4. Conclusion

So, as a result of testing the methods of Fourier analysis and wavelet analysis of cracking of drops during drying or laser drying on MALDI chips, it was shown that such methods give very well distinguishable pictures of qualitative differences between drops. This allows us to speak about the applicability of this method as a method of qualimetry in analytical chemistry and thesiography of biological fluids with MALDI analysis.

Conflict of interest

The authors declare no conflict of interest.

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