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Zipf's Law in Photoacoustics, in the Nature and in the Society

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ABSTRACT

Regularities in some complex systems can sometimes be expressed in terms of simple laws. Some peculiar regularities are identified concerning photoacoustic (optoacoustic) phenomena. In particular, the acoustic signals following phase transitions in liquid irradiated by laser pulses are distributed by magnitude according to the Zipf's law. This power law distribution describes many man made and naturally occurring phenomena, including city sizes, incomes, word frequencies, and earthquake magnitudes. This law suggests connection with anomalous decay, i.e. it implies that small occurrences are extremely common, whereas large instances are extremely rare.
We use this law for signal processing in the course of optoacoustic diagnostics of diluted suspensions. The irradiation of an inhomogeneous liquid sample with a long train of short laser pulses and subsequent recording of a histogram of the magnitudes of the acoustic responses can serve as a diagnostic tool for various applications. The absorption of an incident light by a suspended particle may cause a cavitation event. The random cavitation events also obey the Zipf's law, this fact being used for detection of individual particles.

Keywords: *random sound generation, laser induced cavitation, Monte Carlo simulation*

1. INTRODUCTION

Zipf's law is the observation that frequency of occurrence of some event as a function of the rank when the rank is determined by the above frequency of occurrence, is a power-law function with the exponent close to unity. George Kingsley Zipf (1902-1950), a Harvard linguistics professor, was definitely, not the first who applied decreasing power functions to approximation of sampling distributions. However the results of his research were rather impressive and he was followed by numerous researchers in various fields. These followers have immortalized his name in the name of the law we are going to speak about. The frequency differential form of the Zipf distribution law can be presented by an expression[1]

$$n(x) = \frac{C}{x^{1+\alpha}}, \quad 0 < \alpha < \infty \tag{1}$$

where *n(x)* is the frequency of occurrence, C and α – are constants. Zipf's law is an asymmetric right-hand distribution with an anomalous long "tail". This means, that there are individual counts in the sample having values sufficiently exceeding sample average. The constant α (often called Zipf's parameter) is the very one which is responsible for length of the tail.
Any sampling statistical distribution has both frequency differential and rank forms. Usually it is not too important what form to choose, if the question is natural sciences. However if the sampling volumes are not great, the rank form is preferable. The rank form assumes that instead of a grouping the data they are ranged in such a manner that the first place is awarded to the a unit with the greatest value of the attribute, following is a unit which is exceeded only by the first-rank unit and so on. The rank form is introduced as follows:

$$x(r) = \frac{A}{(r+B)^{1/\alpha}}, \tag{2}$$

where r – is a rank of each of the unit of population according to its individual value x, A and B - constants. The rank form of the Zipf's law distribution (2) is also called a Mandelbrot's distribution[2,3].

Growing interest to the Zipf's law is explained by the fact that its structural features are observed on distribution of many parameters of biological, information, social, economic and other complex systems[2-4].
The known skeptical attitude to some of Zipf's ideas (as it was pointed out by B.Mandelbrot[3], Zipf sometimes tried to find universal laws in those fields where they actually were not present) should not prevent us to use his heritage as a convenient analytical tool for the records' interpretation while doing optoacoustic (OA) diagnostics of the various inhomogeneous liquid media.

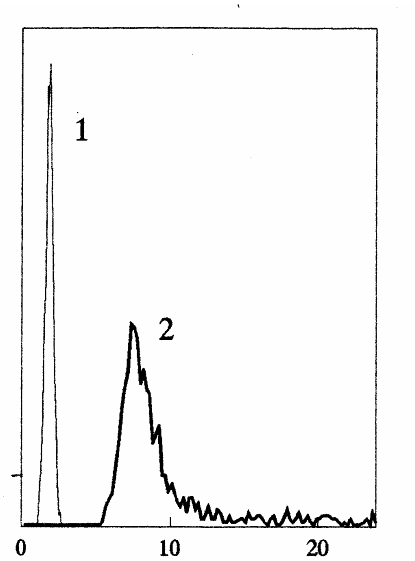
2. PROBABILISTIC ISSUES OF OPTOACOUSTIC CONVERSION

A researcher of a sound generation by laser radiation sometimes comes up against the probabilistic nature of the effect. For example, there were conducted studies of the random acoustic field generated in the sea by the laser irradiation of a wavy surface. Also, the contribution to the overall acoustic signal from a random constituent irradiated by an ensemble of gas bubbles which are always present in the subsurface sea layer and hence distort the optoacoust signal pattern predicted by simplified theoretical models. However the most prominent example of the statistical nature of optoacoustic conversion is a so-called effect of optoacoustic cavitation upon a pulse laser irradiation of the liquid sample containing insoluble absorbing particles. The probe itself is almost transparent for radiation, however the particles are heated by the laser pulse. Then, they deliver energy to the host liquid, the energy value sometimes is sufficient to launch cavitation process which in turn serves as the source of a powerful sound.
First studies of the probabilistic issues of optoacoustic conversion in the low absorbing inhomogeneous liquids dates from the beginning of the Nineties. As it was already well known by this time, OA conversion allowed to achieve the record sensitivity of detection of small amount of impurities in both biological and medical solutions and suspensions. However the instability of magnitude of the acoustic response from one laser shot to another sacrificed all theoretical advantages of a method. A nature of this instability as well as mechanisms, inducing it, called for special research. So let us concentrate on the following OA conversion geometry.
The interaction of a laser beam with weakly absorbing inhomogeneous liquid containing absorbing suspended particles gives rise to optoacoustic conversion within the pencil-shape irradiated volume. Hence the outgoing acoustic signal is usually recorded in the lateral direction outside of the illuminated volume. At low fluence, the optoacoustic signal consists of the thermoacoustic response of the heated host liquid and as well as of heated particles. If a definite fluence threshold condition is satisfied, the temperature of the particles can exceed the boiling temperature of the host liquid. A vapor layer appears adjacent to the particle, which experiences rapid expansion thus giving rise to an effective acoustic signal. The overall acoustic signal obtains so-called cavitation constituent.
As a result, the observed signal is marked by magnitude and waveform both having a random character since the overall signal is composed of the pulses coming from different points and having other random parameters. So, it became recognized, that optoacoustic conversion in low absorption liquids gives rise to so-called combinatory conversion mechanism comprised of a regular response from a host liquid and random signals from cavitation events launched by overheated particles.
Hence, the classical formula for the magnitude of outgoing acoustical response put into the basis of optoacoustic spectroscopy[5]

$$p_m \approx (\mu \beta c^2 E)/(\pi a_0^{3/2} c_p r^{1/2}), \tag{3}$$

applied to the low absorption suspensions, at best describes only the regular constituent of the overall response. Due to the sufficient variations of acoustic response magnitude from one laser shot to another this formula may become useless (here μ is optical absorption coefficient, β is thermal expansion coefficient, c –is sound velocity, E is laser pulse energy, a_0 is the beam waist, r is the broadside distance from the beam axis to the point of observation, c_p is the specific heat of the host liquid). The random process calls for examination of the energy threshold which this or that particle should exceed to launch an individual cavity growth.
The simplest observation of the phenomenon discussed can be provided as follows. The laser beam from XeCl laser was focused in a sample representing distilled twice filtered water. The sample is contained in a cell having UV quartz windows. After filtering procedures only particles less than 0.3 um in diameter remained suspended in the sample. The samples were irradiated by laser pulses at a rate of 0.2 Hz. Each series included 1000 shots at a predetermined energy each. The laser energy was increased from one series to another. The in-phase pickup of these random signals is possible if a relatively low frequency (for example, up to 200 kHz) transducer positioned out of the irradiated "pencil-

shape" volume. The first peak detection of the acoustic signal is obtained by an appropriate gating of the incoming response. After recording the acoustic responses generated by many subsequent laser shots, one can build a histogram of acoustic magnitudes and use it as a diagnostic tool. The results in the form of two selected histograms are shown in Figure 1.



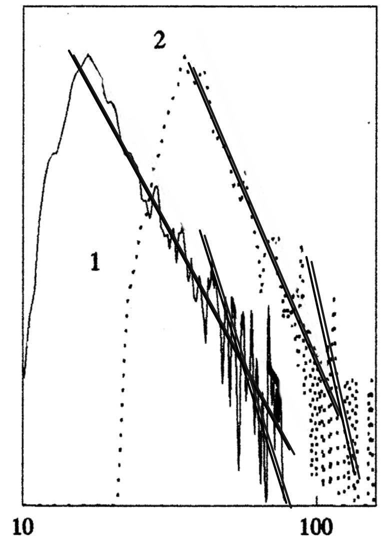


Figure 1. Histograms of amplitudes of the acoustic signals obtained from the irradiation of distilled water with two series of 1000 laser each. The horizontal axis gives the magnitude of the response in Pa, while the vertical one depicts a number of events. The first series corresponds to the fluence 0.08 J/sq.cm (1), while the second is 0.2 J/sq.cm (2). So, the first series corresponds to pure thermoacoustic conversion and the most probable magnitude is well defined by Equation (3). The second series shows the combinatory mechanism of OA conversion the signal having strong cavitation constituent.

Figure 2. The sample is a fresh tap water. It contains a lot of microscopic bubbles, cavitation nuclei and suspended particles. There are shown log-log magnitude histograms the acoustic signals from an irradiation of the probe by two series of 1000 laser pulses each. The first series corresponds to the fluence 0.36 J/sq.cm (1), while the second is 0.45 J/sq.cm (2). Each right slope is well approximated with a pair of inclined straight lines, thus forming Zipf's "knee"[6].

It is for some reason generally believed that if OA conversion is accompanied by phase transitions the signal instability is inevitable. However the experiments conducted at large laser fluence values yield that signal becomes stable again. It turns out, that instability of the response is an attribute of only limited laser energy range.

The histograms obtained in this range, are rather characteristic - the most probable value of parameter (a maximum of the histogram) is displaced to its left edge, while the right part is presented by an abnormally long "tail". It is very similar to Zipf's distribution. However we should investigate this with the use of criteria more rigorous than a simple artistic perception. Besides, it is very important to describe conversion of the histogram with growth of laser energy. Also, we should define the energy value which is necessary to reach in order to return the parameters of conversion under a conventional Gaussian law.

Next, we are going to give the brief review of statistic analysis methods used by experts in the field of Zipf's law. Than we should use some of their experience while conducting Monte Carlo simulation related to the problem we are discussing in the paper.

3. ZIPF’S LAW AS A TOOL. ZIPF’S LAW MANIFESTATION IN THE NATURE AND IN THE SOCIETY

Recently the rank from representation "of long-tail" distributions began to penetrate into natural-science disciplines. One can mention for example, the distribution of elements in the Earth's crust by weight, asteroids in the asteroid belt by mass, distribution of cosmic rays by energy. Some other sources consider distribution of percolation processes.

Among recent papers let us mention[7]. The paper deals with the application of the Zipf’s law to description of the distribution of hot nuclei clasters by their weight and charge for ^{129}Xe. The rank approach is applied for definition of temperature of phase transition of the system. The parameter α appeared to be very informative and convenient tool. With the growth of temperature of the system from 3 up to 7 MeV the tail of distribution was extended, value α decreased from 5.77 to 0.56, and the point of phase transition was found out at the value predicted by model, i.e. at $\alpha =1$. It is no wonder from the point of view of the Zipf’s law architecture because just at this value α the first moment of distribution ceases to converge. It is an indirect indication of transition of the system to less ordered condition.

Zipf’s law is much more popular in the society-based disciplines. However the physicists (namely, the experts in nonlinear dynamics) are also interested in Zipf’s law manifestation in the society and ecology fields. Here are just few examples of papers published in physics journals dealing with various kinds of Zipf-like distributions: distribution of the companies and firms by size[8], distribution of soccer shooters by scored goals[9], distribution of infringers of road rules by the size of penalties[10]. Also, the physicsl journals discuss other specific problems of the humanities, for example, Zipf strategy of investments[11], Zipf’s distribution of citations in scientific literature[12].

The typical Zipf’s distributions relating the humanities, are basically characterized by small values α, approximately in half of all cases $\alpha<1.5$ while $\alpha>10$ is only in 5 % of all cases met. The distributions with small values α (i.e. those having long tails) describe complex hierarchical systems. Small values α define all distributions in linguistics. For example, for both Russian and English languages the distribution of words on their popularity is defined by $\alpha\approx1$[1].

Less flat tails are typical for the distributions relating to the phenomena specific for the Internet and other information technologies. For example, the distribution of web-sites on visitors number, the distribution on web-sites by number of pages contained, the distribution on pages by number links etc[13]. This group is determined by a range $1<\alpha<2$.

The advanced hierarchy systems are the systems of individuals with essentially differing abilities. Such are the distinctions in the intellectual ability, revealed by various tests using open scales[14]. Such are the distinctions in scientific productivity of the scientists determined by well-known scientometric methods[3,12]. The scientists comprise very particular group of almost equally educated people, mostly working in similar conditions. Seemingly, one would expect they will have output distributed about the Gaussian curve around some mean number of papers published. Nothing of the kind! The number of papers varies greatly from one scientist to another. The overwhelming majority of authors do not go beyond a single paper. And vice versa, there are some few authors with an extraordinary paper productivity (several hundred per career). The distinction of scientific articles with respect to their citations is also huge. Typical scientometric distributions are characterized by values $1<\alpha\leq 3$.

Shorter tails mark distributions relating to economy and demography. Such distributions have values $1.5<\alpha\leq5$. For example, these are distributions of people by income, cities by population[15] and others.

Zipf’s law is less popular in physiology, anthropometry and similar disciplines because the distinctions of individuals with respect to their height, weight or reproductive potential are not so great, as distinctions in mental abilities. If met, the Zipf distribution here has a short tail with inevitability. Some of the papers refer to the famous example of distribution of the divorced Italian women by a number of their children. The analysis is carried out on a file of 1718000 women, 500 women having 19 children each, 1500 - 18 children each and so on. The value of determining parameter is $\alpha\approx8$.

This very brief journey into the history of Zipf’s law applications to various fields of knowledge, shows, that a standard practice of the analysis of the distributions which are close to Zipf’s law is revealing conditions at which the system is characterized by $\alpha=1$ (also the cases $\alpha=2$ and $\alpha \gg 1$ are often considered). For example if the Zipf’s parameter reaches 1, there appears a convergence of the first central momentum of distribution. In turn this allows to draw a conclusion about transition of this or that system to more ordered condition.

4. APPLYING ZIPF'S LAW TO OPTOACOUSTIC DIAGNOSTICS OF INHOMOGENEOUS LIQUID MEDIA

To become certain that the tail of histograms fits Zipf's law it is necessary to plot them in log-log coordinate system. Then, the tail should take the form of inclined straight line according to Equation (1) its slope being equal to arctan($1+\alpha$). This can be seen from two moderate fluence series depicted at Figure 2. Such long-tail distributions are usually marked with the absence of the first-order statistical momentum. That should be taken into account while processing the data from OA experiments. It is evident that fluence growth leads to the slope steeping. Finally, at well-above threshold conditions $\alpha\to\infty$ and histogram returns to the Gaussian form (not shown).

It seems, flat distribution tails observed when doing optoacoustics of suspensions can be attributed as to a fractal nature of phase conversions in disperse media[16] and also to certain fractal properties of wave processes[17]. So, the non-Gaussian distribution of acoustic pulses by magnitude is probably related to a fractal nature of optical breakdown in the suspensions having the tendency to clasters' formation. In particular, it is known, that fractal effects in radiation can take place already in case of an ensemble of independent point radiators with fractal spatial distribution (it was just the case of low energy experiment carried out). Let us turn from quality considerations to quantity analysis of the phenomenon.

5. MONTE CARLO SIMULATION OF THE CAVITATION CONSTITUENT

The evaporation threshold depends greatly on the material of the particle. This fact presents the possibility to investigate suspension by means of optoacoustics. Particles of high absorption and high effusivity such as metal particles are of special interest for our study and are considered here in more detail.

In order to simplify estimates within the assumption of independent and single scattering in the medium, we deal with the dilute solutions[18] also we consider the host liquid as a non-absorbing medium. If ε is the laser fluence at the arbitrary point of the beam cross-section, then $\varepsilon\sigma$ is the energy absorbed by a particle which occasionally finds itself at this very point. Here σ is the absorption cross-section of a particle,

$$\sigma \approx \pi\xi d^2/4, \tag{4}$$

where d is the diameter of the particle, and ξ is a coefficient[19], depending on the refraction index of a particle as well as on the ratio d/λ, where λ is an optical wavelength. The absorbed energy causes heating of a particle with subsequent heating and evaporating of the adjacent water layer. Since the effusivity of metal is much higher than that of water, it is natural that a major fraction of the energy absorbed by a particle is spent to heat itself. Hence, the energy conservation equation for threshold fluence ε_{th} is

$$\varepsilon_{th}\sigma \approx \pi\rho C d^3 \Delta T/6, \tag{5}$$

where ρ is the density of particle, C is a specific heat of the particle, ΔT=80ºC. As a result we get for the threshold fluence for these type of the particles;

$$\varepsilon_{th} \approx 2\rho C d \Delta T/3\xi, \tag{6}$$

that yields $\varepsilon_{th} \sim d$, if ξ is independent of d that is valid for large particles, $d>\lambda$, or, alternativaley for small particles of some specific kind. Such particles, for example, can be gold nanoparticles, now popular in medical diagnostic practice. In the further simulation we just consider the suspension made of such particles. At simulation it was accepted, that:

- The amplitude of a resulting signal is represented by the sum of amplitudes of signals from each particle which have absorbed energy with a value exceeding the threshold one. "Underthreshold" particles do not radiate any sound.
- The amplitude of a signal, radiated by an overthreshold particle p is defined by an expression $p \sim d^2(\varepsilon - \varepsilon_{th})$[20]. All the particles have the same diameter d, i.e. we deal with monodisperse suspension.
- The total number of particles in a sample was 100. They have random positions in the probe. Only a small share of the ensemble is irradiated by a laser beam. According to ε_{th} for the given diameter of particles the normalized fluence in the focal waist ε_0 is introduced which is the key simulation parameter. For example, ε_0 =1 means, that

fluence in the focal waist is equal to ε_{th} (the energy value sufficient for heating the given particle to 80C and achieving the boiling threshold);

- The second basic parameter is the focal area diameter. It is set equal to 1, other geometrical parameters being related to it. So, the length of focal area is equal to 30.
- The space where the OA conversion takes place and where 100 particles find themselves in random positions changing from one laser pulse to another is represented by a cylinder coaxially enveloping the laser beam. The length of the cylinder expressed in focal area diameter units is equal to 6000, its cross-section diameter is equal to 6.

The simulation included Monte Carlo procedure for 7 series each having 200000 trials. The fluence ε_0 was maintained constant within each series. It took seven values changing from one series to another, i.e $\varepsilon_0 = 1.1; 1.3; 2; 5; 10; 100, 10^5$. Records of series were used for construction of histograms, as well as of rank distributions, for definition of mean sample magnitudes and other statistical characteristics of OA conversion.

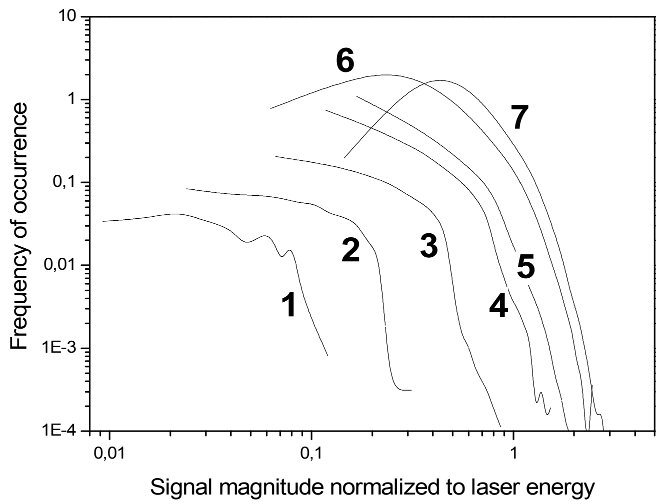


Figure 3. Model log-log histograms of magnitudes for a set of laser fluence values ε_0 =1.1 (curve 1); 1.3 (2); 2 (3); 5 (4); 10 (5); 100 (6), 10^5 (curve 7)

The results of the simulation according to this simplified model are presented in Figures 3-5. Figure 3 shows the histograms distributed over 20 bins each. It appears that 5 first series have the most probable (modal) acoustic signal magnitude value equal to zero (not shown). The log-log histograms for series 1-5 also exhibit knees typical for Zipf's distributions. Within small magnitude range the curves are rather flat corresponding to Zipf's parameter $\alpha\approx 1$ of the Equation (1). On the contrary, within large magnitude range the histograms are steep, i.e $\alpha \gg 1$. The last two series (curves 6 and 7) show the principal phenomena, i.e. the formation of nonzero modal magnitude.

To get the simplified estimate of α as a function of ε_0 there were plotted rank magnitude distributions for the same series (Figure 4). Now, the curves can be approximated by the Equation (2) and as opposed to Figure 3, the large magnitude range occupies the left part of the plot (that means that all 200000 scores for each series are sorted according to their magnitudes). The slope of each auxiliary chord is approximately equal to $1/\alpha$.

Figure 5 shows fluence dependencies of various optoacoustic conversion parameters as derived from the simulation. The shape of a curve 1 (mean sample magnitude) is close to the simplest prediction of papers[5,21].

Let us point out that curve 1 and curve 2 (median magnitude) converge with the fluence growth. This is an indication that optoacoustic conversion parameters turn to good repeatability. The growth of $\alpha(\varepsilon_0)$ from 5 to 10 also points to this tendency. As we know, at $\alpha\approx 1$ Zipf's distribution is almost no different from Gaussian law.

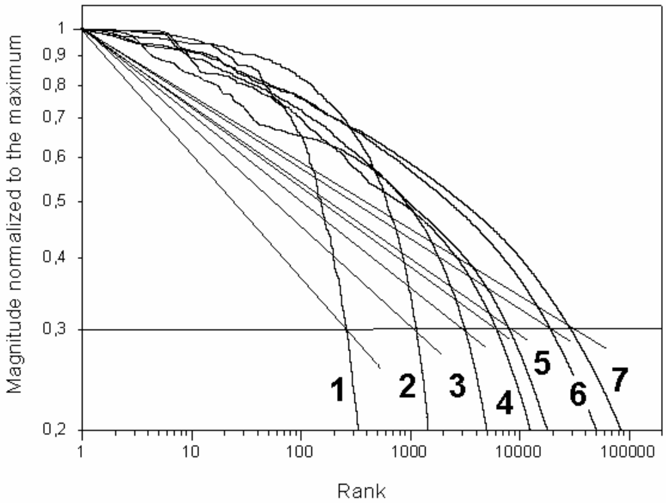


Figure 4. Model log-log rank magnitude distribution for the same set of laser fluence values ε_0. The purpose that is served by a set of tilted straight lines is the determination of average slope of each curve within the range of largest magnitudes (at the $1/e$ level, horizontal straight line). Then, the slope thus derived is used to calculate the Zipf's parameter α.

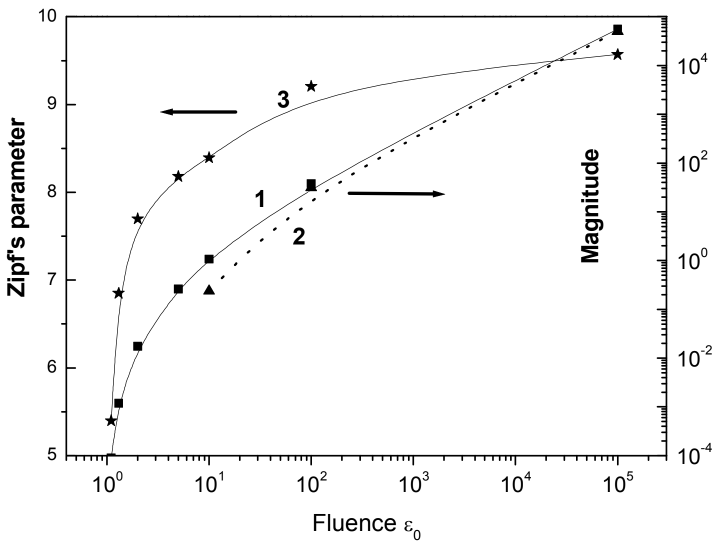


Figure 5. Dimensionless acoustic signal magnitude vs laser fluence (right vertical axis). Mean magnitude (curve 1) and median magnitude (curve 2) are shown. Zipf's parameter α vs laser fluence (curve 3) is layed off along the left Y-axis.

6. CONCLUSIONS

The simplified Monte Carlo simulation, in general, confirmed the experimental observationof the Zipf's law signal magnitude distribution within the moderate energy fluence range in the course of optoacoustic conversion in inhomogeneous liquids. The previous experience of application of Zipf's statistics to various fields of knowledge helps to restore the statistical parameters of OA conversion.

Other important conclusion consists in necessity of taking into account the mean sample acoustic magnitude vs laser energy in the case when fluence only insignificantly exceeds the fluence of pure thremoacoustic conversion. The magnitude histograms have especially long tails, and, hence, α<1. As we now know, this means that mean sample acoustic magnitude depends on the sample volume, i.e. it does not converge to any finite quantity as it is typical for Gaussian law. This unstable character of mean magnitude, apparently, explains the contradictions between different papers of the Eighties and Nineties dealing with acoustic response magnitude p_m as a function of laser energy input E. The papers presented a variety of experimental curves of $p_m(E)$, not consistent with each other, power law approximation yielding power from 5/6 up to 4).

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