Laser Physics, Vol. 10, No. 6, 2000, pp. 1293–1297.

Controlling the Radioactive Decay of Long-Lived Mössbauer Isomers

S. K. Godovikov

D.V. Skobeltsyn Institute of Nuclear Physics, M.V. Lomonosov Moscow State University, Vorob'evy gory, Moscow, 119899 Russia

e-mail: blokh@srdlan.npi.msu.su

Received June 15, 2000

Abstract—The possibilities of controlling the radioactive decay of long-lived isomers with intermediate Mössbauer levels (^{119m}Sn, ^{125m}Te) up to complete decay suppression and γ-ray emission are experimentally demonstrated. A physical model of this phenomenon based on the concept of "retarded" nuclear states produced under the action of standing Mössbauer waves is proposed. The way to implement an instantaneous flash of coherent γ-ray emission within the energy range of several tens of kiloelectronvolts is discussed.

1. INTRODUCTION

The possibilities to control the radioactive decay of long-lived (~10^2 days) isomers with Mössbauer intermediate levels have been experimentally demonstrated for the first time in a series of papers [1–6], which were published in 1998–1999. The example of ^{119m}Sn isotopes (the energy of the isomer level is 89.53 keV, $T_{1/2}$ = 293 days, and the energy of the Mössbauer level is 23.87 keV) was used to show that a closely spaced Mössbauer screen allows the decay to be decelerated down to $\Delta\lambda/\lambda = -(0.114 \pm 0.027)$, where λ is the decay constant. Driving the nuclear level by varying $\Delta\lambda/\lambda$ within the interval from –1 (decay suppression) to +∞ (a single flash) is a crucial problem for the creation of a γ-ray laser. Preliminary experiments with a superscreen, where ^{119}Sn and ^{119m}Sn isotopes are mixed on the atomic level have shown that a variation in $\Delta\lambda/\lambda$ may be even larger.

Three main principles were used to interpret the discovered phenomenon: (1) a screen and a source produce a system of standing waves, and nuclei in a predecay state may be located around the antinodes of these waves; (2) the isomer level of a predecay-state nucleus and an antinode of the standing wave form a system of two oscillators whose vibrations are coupled through dynamic synchronization (the nuclear Huygens effect); (3) this interaction abruptly changes parameters of the isomer level. This interpretation was used as a basis for a prediction that, in certain situations, the decay curve may oscillate with a macroscopic period of time.

This paper presents the results of further experimental and theoretical studies devoted to the control of the radioactive decay of long-lived isomers with Mössbauer energy levels (^{119m}Sn, ^{125m}Te).

2. EXPERIMENTAL TECHNIQUE

Experiments were performed in three different directions.

(1) The first direction included investigations of a "new" source placed inside a Mössbauer screen (with a geometry of a 4π solid angle). A Ba^{119m}SnO$_3$ Mössbauer source with an activity of 3 mCi produced in 1999 was employed in these experiments. The content of ^{119}Sn isotopes in this source was negligibly small, i.e., the inner screen was missing. The source had a shape of a ∅10-mm thin disc. The screens had a shape of ∅14-mm discs containing a black-body Mössbauer absorber [4]. The content of stable isotopes in the screens was very high (15 mg/cm^2 of ^{119}Sn in screen no. 1 and 24.4 mg/cm^2 in screen no. 2). The screen no. 1 was removed during the measurements. Both screens were removed in test experiments.

(2) The second direction included studies of an "old" Ca^{119m}SnO$_3$ source, which was produced in 1989 and which had an initial activity of 50 mCi. This source contained ~2000 stable atoms per each radioactive atom (the activity was ~25 μCi) by the beginning of our experiments (March 11, 1998). These stable atoms served as a resonance screen separated by subatomic distances from emitting nuclei. The analysis of the amplitude emission spectrum of this source performed with the use of a Si(Li) detector has demonstrated the absence of impurities of other isotopes (except for ^{125}Sb, whose content did not exceed several percent).

(3) The third direction included investigations of an "old" Ca^{119m}SnO$_3$ source, which was produced in 1987. Virtually no emission of ^{119m}Sn isotopes was observed for this source, while the emission of the ^{125}Sb(^{125m}Te) impurity with an activity of 25 μCi played a dominant role by the beginning of our experiments (March 11, 1998). The decay scheme of ^{125}Sb isotopes is shown in

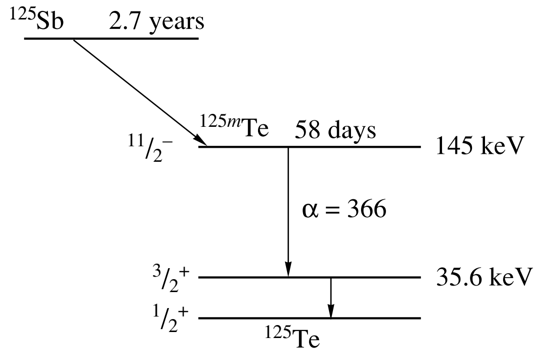

Fig. 1. Diagram of decay of a ^{125}Sb isotope (α is the conversion coefficient).

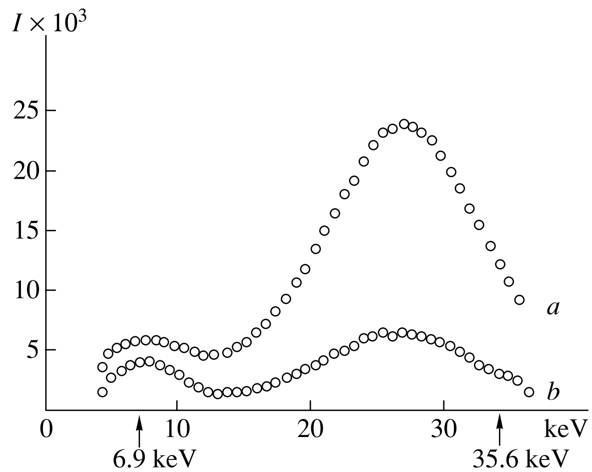

Fig. 2. Emission spectrum of ^{125}Sb (^{125m}Te): (*a*) in the absence of a filter and (*b*) with a copper filter with a thickness of 50 μm.

Fig. 1. The decay of ^{125}Sb is rather slow ($T_{1/2}$ = 2.7 years and $\lambda = 70.16 \times 10^{-5}$ 1/day). Conversely, the decay of the derivative isotope ^{125m}Te is rather fast ($T_{1/2}$ = 58 days). A circular equilibrium between ^{125m}Te and ^{125}Sb is achieved within the first year of the source lifetime. After that, the production rate of ^{125m}Te isotopes is equal to the decay rate of parent isotopes. Figure 2 displays the amplitude emission spectrum of the source measured with the use of a NaJ(Tl) crystal with a thickness of 0.1 mm. The left-hand peak in this spectrum is the "ejection peak" with an energy of 6.9 keV, while the right-hand peak corresponds to a combination of Te and Sb X-ray emission (K_αTe = 27.4 keV and K_αSb = 26.3 keV) and nuclear emission of ^{125m}Te with 35.6 keV. Using a copper filter with a thickness of 50 μm, we were able to considerably suppress the X-ray background in the right-hand peak. The ratio of the intensities of the left- and right-hand peaks drastically changes in such a situation (Fig. 1b), which indicates that the left-hand peak is due to the action of nuclear emission of ^{125m}Te (6.9 keV = 35.6 keV ^{125m}Te – 28.7 keV $K_\alpha J$) on a NaJ crystal. Emission of ^{119m}Sn isotopes (23.8 keV, 25.2 K_αSn) cannot give rise to any ejection "peak."

Precise λ measurements in all the three cases were employed with the use of the scintillation detection technique (a NaJ crystal with a thickness of 1 mm) and a multichannel analyzer operating in the multiscalar regime. The pulses were distributed within an energy window of the corresponding nuclear (Mössbauer) line in 256 channels in the regime of time sweep with a frequency of 40 Hz (100 μs per channel). The source intensity was measured from the time corresponding to 2^{16} counts per channel. This time typically falls within the range of hours. The error of measurements was mainly associated with the error of determining the moment of time when the level of 2^{16} is achieved. The count rate J, i.e., the number of pulses per second per channel, is a measurable parameter under these conditions.

The distance between the source and the detector in the first series of experiments was about 13 cm. In the second and third series of experiments, the sources were fixed on the housing of the emission detector. A Pd filter with a thickness of 50 μm, suppressing the accompanying X-ray emission, was employed in all the series of measurements.

3. EXPERIMENTAL RESULTS

Series no. 1. The results of studies devoted to decay deceleration with the use of screens in the 4π geometry are presented in Fig. 3. Measurements were performed during ~150 days in both real experiments (*a*) and testing measurements with removed screens (*b*). For the convenience of comparison the lines *a* and *b* are given with the same time reference point. As is seen from the figure, dependences *a* and *b* can be satisfactorily approximated with exponential functions. In other words, these dependences are represented by nonparallel straight lines in the logarithmic scale (the dashed line in Fig. 3 is parallel to line *b*). Processing these dependences with the least-squares method, we find that the decay constant corresponding to line *b* is equal to $(236 \pm 5) \times 10^{-5}$ 1/day, which is normal. In the case of line *a*, the decay constant is reduced down to $\lambda = (177 \pm 7) \times 10^{-5}$ 1/day, which corresponds to $T_{1/2}$ = 391 ± 15 days, i.e., $\Delta\lambda/\lambda = -(0.25 \pm 0.03)$. This value of $\Delta\lambda/\lambda$ is two times larger than the value of this ratio measured in [3, 4], which indicates the efficiency of using the geometry of complete source screening.

Series no. 2. The decay of an "old" $Ca^{119m}SnO_3$ source is illustrated in Fig. 4. Measurements of this series were performed during ~640 days. As is seen from these results, the decay in this case can also be satisfactorily approximated with an exponential function with decay parameters equal to $\lambda = (160 \pm 2) \times 10^{-5}$ 1/day and $T_{1/2}$ = 433 ± 5 days. The dashed line corresponds to a normal decay law with $T_{1/2}$ = 293 days. We also find that $\Delta\lambda/\lambda = -(0.32 \pm 0.01)$, i.e., this ratio is even greater than in the case of series no. 1.

Series no. 3. Figure 5 presents the decay curve for an "old" source of ^{125}Sb in a $CaSnO_3$ matrix. We use a conventional nonlogarithmic ordinate axis in this case. Measurements were carried out in parallel with experiments of series no. 2 and during the same days as experiments of series no. 2 within a period of time of ~800 days. The dashed line in Fig. 5 shows the normal law of ^{125}Sb decay with $T_{1/2}$ = 2.7 years. The behavior of the curve in Fig. 5 considerably differs from an exponential behavior, featuring oscillations with a period of ~220 days. The activity becomes higher than the initial value, i.e., we deal with radiation generation, at three points (corresponding to 220, 440, and 660 days). A short-period fine structure (local maxima and minima of radiation intensity) is also observed around the last point.

Such an unusual behavior of the radioactive decay curve for a separate isotope is observed for the first time since the discovery of radioactivity.

4. A PHYSICAL MODEL AND DISCUSSION OF THE RESULTS

The results presented above and published in [1–6] allow us to propose a physical model of the considered processes, which permits a more efficient planning of further experiments and provides a background for a more detailed interpretation of the processes under study.

Our main concept of controlling the process of radioactive decay is to include a nucleus that still undergoes decay into a system having an additional excited energy level. This problem can be easily solved by placing a nucleus at an antinode of a standing wave produced by radiation characteristic of a given isotope. A γ-emission standing wave can be excited only under conditions of the Mössbauer effect, which allows emission and scattering processes to be implemented without a recoil and which ensures the coherence of the scattering processes, giving rise to a stable interference pattern with a lifetime of ~10^{-8} s. This interaction time, which is huge on the intranuclear scale (10^{-23} s), results in the appearance of a nucleus–standing wave system with two excited energy levels. In other words, a system of two coupled oscillators with multiple (for the considered class of isotopes) or virtually multiple eigenfrequencies arises. As is well known from mechanics, radio physics, and optics, the so-called Huygens effect, or the dynamic synchronization of vibrations in frequency and phase, may occur under these conditions. The upper nuclear level may change its parameters in some way. This approach does not require high-power radiation fluxes, high energies, etc. It would be sufficient to employ such a tool as coherent emission inherent in the nucleus itself to finely modify a system of intranuclear processes.

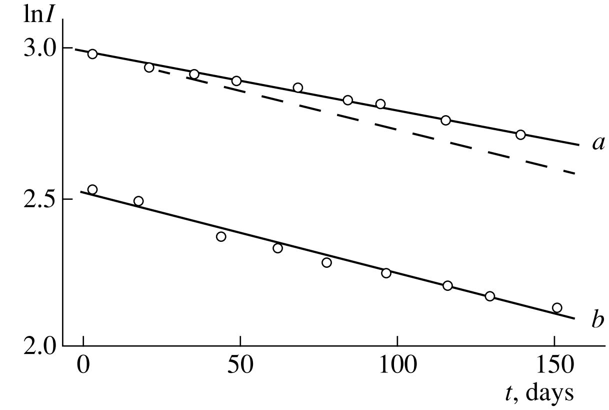

Fig. 3. Decay curves of a $Ba^{119m}SnO_3$ source (the error is within the circle): (a) with a screen in the geometry of a 4π solid angle and (b) without a screen.

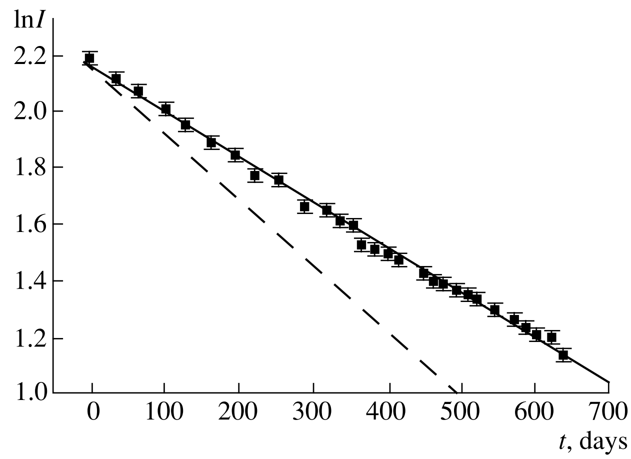

Fig. 4. Decay curve of an old $Ca^{119m}SnO_3$ source with an effect of inner screening.

What is the result of the considered process of vibration synchronization? This result is quite unexpected. The predecay-state nucleus under these conditions is transferred to some new state where the half-decay period is indefinitely large. This nucleus is "retarded," being out of the radioactive decay for a while. The further evolution of such nuclei may be also very unusual.

Let us try to find some mathematical representation for the above-introduced physical concepts. It is quite clear that the standard equation of radioactive decay,

$$dN/dt = -\lambda N, \qquad (1)$$

where N is the number of radioactive nuclei at a given moment of time, requires some modification to include the above-discussed factors. Suppose that the number of nuclei per unit source volume emitting at a given moment of time is equal to λN. The formation of a standing wave is a typical example of pair interaction between an emitting nucleus and a scattering nucleus. As is well known, the probability of such processes is proportional to the square of the concentration of com-

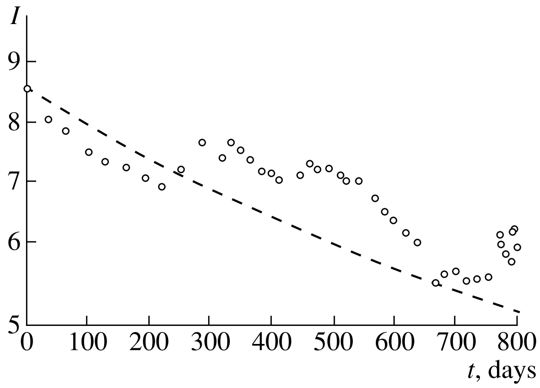

Fig. 5. Decay curve of an old $^{125}Sb(^{125m}Te)$ source in a $CaSnO_3$ matrix (the error is within the circle).

ponents, involved in the interaction, i.e., $\sim(\lambda N)^2$. We assume, of course, that the number of scattering nuclei is not less than λN, which is always true for the considered experiments. Consequently, the number of "retarded" nuclei is given by a product of the probability that a system of standing waves appears and the number of nuclei N that did not undergo decay, i.e., $\sim(\lambda N)^2 N$. In other words, this number is equal to AN^3, where A is some proportionality constant. Retarded nuclei are no longer involved in the decay process. Therefore, Eq. (1) should be rewritten as

$$dN/dt = -\lambda(N - AN^3) = -\lambda(1 - AN^2)N. \quad (2)$$

As can be seen from Eq. (2), the equation of radioactive decay becomes essentially nonlinear. Analysis of Eq. (2) brings us to the following conclusions:

(1) Since $(N - AN^3) < N$, i.e., $(1 - AN^2) < 1$, the absolute value of $\lambda(1 - AN^2)$ is less than λ, leading to decay deceleration;

(2) The quantity N decreases in the course of time, and $\lambda(1 - AN^2) \longrightarrow \lambda$, i.e., the decay deceleration effect gradually vanishes;

(3) The decay can be completely suppressed, with $dN/dt = 0$, for some relation between A and N. In accordance with Eq. (2), such an effect may occur if

$$N = \pm\sqrt{1/A} \quad (3)$$

For example, we have $N \approx 6.77 \times 10^{15}$ for a source with an activity of 5 mCi, which requires that $A \approx 2.18 \times 10^{-32}$.

Calculated values of the parameter A

Series number	Activity of the source	Isotope	A
Series of (original) studies [3, 4]	5 mCi	^{119m}Sn	2.5×10^{-33}
no. 1	3 mCi	^{119m}Sn	15.1×10^{-33}
no. 2	25 μCi	^{119m}Sn	2.8×10^{-28}
no. 3	25 μCi	^{125m}Te	0.783×10^{-28}

If A becomes larger than this quantity for some reason, then condition (3) can be also satisfied for weak sources;

(4) Condition (3) implies the bifurcation of the decay process. Two scenarios are possible above the point defined by Eq. (3) for given A: the plus sign corresponds to a conventional decay, while, in the case of the minus sign, Eq. (1) can be rewritten as

$$dN/dt = +\lambda N, \quad (4)$$

i.e., N exponentially grows with time. Such a process is possible if the nuclei retarded at earlier moments of time are transferred to the active state because of some reason, which is equivalent to the introduction of a flux of "new" nuclei into the system and radiation "generation";

(5) Apparently, the subsystem of retarded nuclei transferred to the normal state is characterized by its own decay constant λ. In principle, this constant may exceed the initial decay constant, i.e., $\Delta\lambda/\lambda > 0$. This is possible due to the Le Chatelier–Brown principle, which implies that an external action driving a system out of the equilibrium position gives rise to processes in the system that tend to attenuate the result of this action;

(6) The branches corresponding to plus and minus signs in Eq. (3) are energetically equivalent, which means that decay and generation processes may replace each other, i.e., oscillations may arise in the dependence of N on t. The period of these oscillations should be a macroscopic quantity, because the processes giving rise to the formation of a subsystem of retarded nuclei are slow. Solutions to equations of the form of Eq. (2) are well known in the general theory of self-organizing systems [7].

The results of our analysis illustrate a high efficiency of using Mössbauer screens for controlling the radioactive decay of long-lived isomers with intermediate Mössbauer levels. The parameter A characterizes this efficiency. The data obtained for $\Delta\lambda$ allow us to estimate A. The results of these calculations are summarized in the table. When using the formula

$$\lambda_e = \lambda(1 - AN^2), \quad (5)$$

where λ_e is the experimentally measured decay constant, to calculate A, we employed the values of N corresponding to the initial moment of time in a series of measurements. A situation with $dN/dt = 0$ was implemented in the series no. 3. As can be seen from the table, the use of a screen with the 4π geometry increases the efficiency of radioactive-decay control by a factor of six as compared with the case of a single-side screen. This finding indicates a substantially nonlinear character of the dependence of the parameter A on the amount of stable isotope in the environment of a predecay-state nucleus. Indeed, A catastrophically (by a factor of 10^5) increases in the regime of a superscreen, where radioactive and stable isotopes are mixed on the atomic level (series nos. 2 and 3). Due to the fact that emitting and scattering nuclei are very close to each other in this case, an interference pattern with many standing waves and corresponding antinodes of the electromagnetic field arises in these experiments. Starting with $A \sim 10^{-28}$, decay deceleration is observed even for sources with a very low activity (~25 μCi). As can be seen from the table, the regime with $dN/dt = 0$ cannot be implemented for the initial series, since the parameter A in this case is approximately ten times lower than it is required ($\sim 2.18 \times 10^{-32}$). More favorable conditions are achieved in the series no. 1. However, A is still lower than the required value. In the series no. 2, the critical value is $A \sim 8.7 \times 10^{-28}$, which means that the system is very close to generation. Finally, the series no. 3 demonstrates this state.

Let us consider the data of the series no. 3 in greater detail. We can understand the appearance of oscillations in the decay curve by analyzing Eq. (2). The quantity $\lambda(1 - AN^2)$ remains positive from the moment of time corresponding to the beginning of measurements up to the first point where decay is completely suppressed ($t_1 = 220$ days). Such a situation corresponds to a somewhat decelerated normal decay. This quantity is equal to 0 at the point t_1. For $t > t_1$, this quantity reverses its sign because of a drastic increase in the number of radioactive nuclei in the source. Let us try to provide quantitative estimates for this effect. The first growing section from the level of $J = 6.945$ up to the level of 7.662 (or from 1 up to 1.103 in relative units) is observed in Fig. 5 within 66 days. Solving the equation

$$\exp[70.16 \times 10^{-5}(0.783 \times 10^{-28} N^2 - 1)] = 1.103 \quad (6)$$

we find that $N = 1.99 \times 10^{14}$. At the point t_1, we have $N = 1.13 \times 10^{14}$, i.e., the number of nuclei has increased by a factor of 1.76. What could be the reason for such an abrupt increase in N? About 20 stable ^{125}Te atoms per single radioactive atom have been accumulated in the source from the moment it was produced up to the moment t_1. Apparently, this amount of stable atoms corresponds to a critical threshold where a further growth in this amount results in the collective involving of ^{125m}Te nuclei retarded at earlier moments of time in the decay process. It is quite probable that this process may have some degree of coherence due to its collective nature. The excess of ^{125m}Te nuclei gradually decreases, reaching the value that corresponds to the equality $\lambda(1 - AN^2) = 0$ (the first maximum in the curve shown in Fig. 5). Then, a conventional decay is observed for $\lambda(1 - AN^2) < 1$. This stage lasts until the number of retarded nuclei is compensated up to a threshold value, whereupon the decay stops again, which is followed by the second flash. Then, the cycle is repeated. The duration of the second cycle is also about 220 days, and the ratio $\Delta\lambda/\lambda$ is positive for the decreasing section of the considered curve.

Now, let us consider similarities and differences in the results of experiments with ^{119m}Sn and ^{125m}Te isotopes. Similarities include variations in the decay constant under the action of any type of a Mössbauer screen. The critical thresholds of concentration of a stable isotope in a source where these variations arise are also similar: 20 stable atoms per single radioactive atom. This conclusion follows from the results of preliminary experiments with a $Ca^{119m}SnO_3$ source employed earlier in studies [3, 4] performed in 1999, when a considerable amount of ^{119}Sn was accumulated in the source. These studies have shown that the self-deceleration of decay starts with a threshold of 20 : 1.

An easily noticeable difference is that flashes indicating radiation generation, similar to those observed in Fig. 5, are not detected for ^{119m}Sn. Apparently, this is due to the fact that the efficiency of dynamic synchronization processes for this isotope is lower because the ratio of level energies is not exactly multiple in this case, 89.53 keV/23.87 keV = 3.75, in contrast to ^{125m}Te, where this ratio is 145 keV/35.6 keV = 4.07.

5. CONCLUSION

The proposed physical model and the corresponding interpretation of the observed phenomena allowed us to use a few simple concepts to provide a sufficiently complete description for the entire set of experimental data. The proposed method of controlling the radioactive decay of the considered class of isotopes is quite constructive for physical and practical applications. Indeed, since the generation of γ-emission is observed in the spontaneous regime in a physical object (a Mössbauer source), this process can be enhanced by means of fine radio-chemical technology. In particular, by abruptly increasing the concentration of retarded nuclei, one can produce, in principle, an instantaneous radiation flash, i.e., $-\lambda(1 - AN^2) \longrightarrow +\infty$. Radiation emission would be collective and coherent in this regime. This would be the first practical step toward the creation of an operating γ-ray laser device.

ACKNOWLEDGMENTS

I am grateful to L.A. Rivlin for his interest in the results of this work.

This study was partially supported by the Russian Foundation for Basic Research (project no. 99-02-16357).

REFERENCES

1. Godovikov, S.K., 1998, *9th Conf. on Laser Optics, Technical Program* (St. Petersburg), p. 71.
2. Godovikov, S.K., 1998, *Russian Conf. "Applications of Nuclear-Physical Methods in Magnetism and Science Materials, Abstracts* (Izhevsk), p. 87.
3. Godovikov, S.K., 1998, *JETP Lett.*, **68**, 629.
4. Godovikov, S.K., 1998, *Laser Phys.*, **8**, 1100.
5. Godovikov, S.K., 1999, *Izv. Akad. Nauk, Ser. Fiz.*, **63**, 1397.
6. Godovikov, S.K., 1999, *JETP Lett.*, **70**, 647.
7. Nicolis, G. and Prigogine, I., 1990, *Exploring Complexity* (Moscow: Mir).