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In sæcula sæculorum: A lab activity to create with students a radioactive secular equilibrium model(*)

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Summary. — The teaching of radioactivity in the high school is often difficult to implement, especially from an experimental point of view. In this paper an activity based on a laboratory experiment on radioactivity is presented. The activity was proposed to high school students in their fourth year of studies attending the summer internship organized by the University of Pavia at the Department of Physics. The experiement concerns the radon decay chain, and in particular the measurement of the activity of ^{214}Bi, both in absence and in presence of its progenitor ^{222}Rn. This way it is possible to observe two different decay curves: the first provides a ^{214}Bi decay constant in agreement with the theoretical one, whereas the trend of the second one can be understood only through the hypothesis of secular equilibrium with ^{222}Rn. Using an engaging and interesting game with dice, a model of radioactive decay was developed and performed with students divided in small groups.

1. – Introduction

Among the interesting subjects of modern physics, radioactivity is one of the topical arguments worth to be studied in depth. Within its framework students can acquire ideas and scientific terms useful to approach the debate from a critical point of view [2]. Many interesting educational laboratory activities about the phenomenon of radioactive decay have indeed already been implemented and proposed [3, 4]. In this article, we present a laboratory experiment to study radioactive equilibrium occurring within a natural decay family. In support of this experience a model based on a game with dice has also been developed to focus on the fundamental aspects of the radioactive equilibrium phenomenon. This experience has been presented during the annual summer internship organized by the Department of Physics of the University of Pavia in collaboration with National Institute of Nuclear Physics (INFN) —section of Pavia— and attended by thirty students of the fourth year of secondary school. The students, divided into five groups —each one assisted by a tutor—, spent two weeks into the Department laboratories attending various seminars. They also participated actively in laboratory experiments

(*) From the Master Degree Thesis [1].

concerning several physics topics including the one presented in this work. The model with dice is designed to describe not only the probabilistic feature of the radioactive decay [5] but also to simulate the decay of a radioactive chain. For this reason, we use two sets of chained throws: the dice discarded by the first set are passed to the next set. In this way it is possible to observe how the trend of the second set is dependent on the discard probability of the first one. Consequently, this model is able to simulate the decay of a radioactive family in which an equilibrium is established between its radionuclides. The experiment consists in observing the radioactive decay chain of Radon and especially in measuring the activity of one of its "daughters" (^{214}Bi) both in absence and in presence of the progenitor (^{222}Rn) to highlight —in two different cases— if a radioactive equilibrium is established. Preliminary knowledge required by students for such activity is minimal: the exponential function, the properties of logarithm and some basic notion about radioactive decay.

2. – Model with dice: how to describe the probabilistic aspect of the radioactive decay

To simulate the throw of the dice students made use of the free application "Roll the Dice" which allowed them to throw any number of dice. Two groups of students were formed —hereinafter we will call them group A and group B— in order to simulate initially the independent decay of two different radionuclides. The model is based on the following rules: at each throw group A discards the dice whose result is equal to 1; group B discards the dice whose result is 1, 2, 3 or 4.

This implies that probability associated with group A to discard one die is equal to $1/6(P_A \sim 0.167)$; while the probability of discard for the group B is equal to $4/6(P_B \sim 0.667)$. Students reported the number of discarded dice in function of the number of throws obtaining the respective "discarded laws". Figure 1 shows the graphs realized by the two groups. Students confirmed that the two laws follow an exponential trend using the fit function $y = Ae^{-\lambda x}$. Each group observed that the numerical coefficient of the exponent found in the fit function was quite similar to their own value of the discard probability. Through simple mathematical steps it was possible to demonstrate that this intuition was correct: the numerical coefficient corresponds appoximately to the probability of discarding the dice [6]. This is the fundamental core of the model which allows students to establish a direct analogy with the radioactive decay: so, it is possible to associate the amount of dice to the number of radionuclides, the discard probability to the decay constant and the number of discarded dice to number of radionuclides that decay in a unit of time, *i.e.* the radioactive activity. It is interesting to underline that this model is based on a constant probability phenomenon just like the radioactive decay. In fact, as no one can predict with accuracy how many dice are discarded during each throw, also no one can predict how many radionuclides decay in a unit of time. Finally, this model allows to derive the exponential trend of radioactive decay without complex mathematical derivations (*e.g.* the resolution of a differential equation) which are out of the knowledge of high school students in their fourth year of studies.

3. – Model with dice: how to describe the behaviour of a radioactive decay chain

The model is evolved to simulate the behaviour of a decay chain composed of only two radionuclides: the first (the parent) decays into the second (the daughter) which in

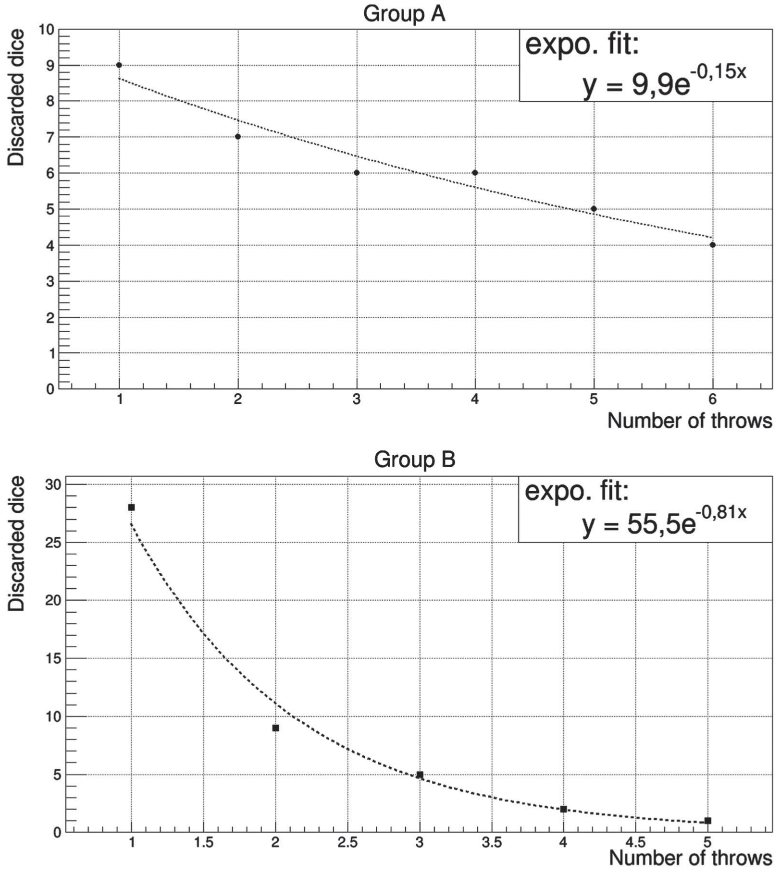

Fig. 1. – Graphs obtained by the two groups (A and B) in the dice model. Each group used an exponential fit function.

its turn decays. Students quickly grasped that, in order to realize such situation, the discarded dice by a group (*e.g.* group A) had to be added to the other group (group B). In this way, group A continued to discard the dice whose result was equal to 1 and passed them to group B, which continued to discard the dice whose result was 1,2,3, or 4. Students recorded the number of discarded and remaining dice at each throw. The collected data are shown in the graph (fig. 2).

Students easily identified an exponential trend and consequently they realized a fit using an exponential function $y = Ae^{-\lambda x}$ as before. In this way group A obtained the following numerical coefficient of the exponent:

$$\lambda_A = 0.17.$$

Therefore, this value was in good agreement with the discarding probability, just like in the previous simulation. Instead, using the same fit function, group B obtained

$$\lambda_B = 0.26.$$

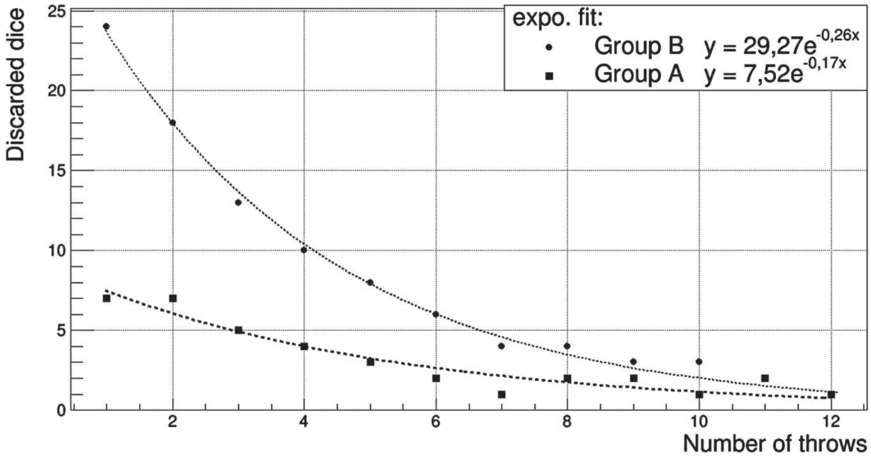

Fig. 2. – Number of discarded dice by the two groups (A and B) in the model used to simulate the behaviour of a decay chain.

This result was quite different from the previous one obtained by group B. In fact, it was not in good agreement with the discard probability of group B (~ 0.67) but it was much closer to the probability of group A (~ 0.17). During the simulation, group B realised that, after some throws, the ratio between the discarded dice and those received from the first group was roughly constant: so a situation of equilibrium was established between the two groups. Under these conditions, it was possible to conclude that the probability of group A influences the number of dice discarded by group B and so the probability associated to group B become gradually more and more similar to the one of group A. This result was then transferred to the case of a real radionuclide chain, which the dice experiment was meant to simulate. Consequently, it was easy to deduce that the daughter activity was governed by the decay constant of the parent. However, this is true only in the examined case in which $P_A < P_B$, which implies that $\lambda_{parent} < \lambda_{daughter}$. By means of this model, students understood that every time this relationship occurs between the decay constants of two radionuclides then the radioactive equilibrium is established: the daughter decays with a decay constant equal to the progenitor's one.

4. – The experiment: measurement of the decay constant of a radionuclide within its radioactive decay chain

The experiment concerns the measurement of the activity of ^{214}Bi within its natural radioactive chain [7-9]. ^{214}Bi has a half-life equal to 19.7 minutes, and it belongs to the natural radioactive family of ^{238}U. However, the measurement is performed to observe ^{214}Bi within a smaller decay chain: from ^{222}Rn onwards. Therefore ^{222}Rn (half-life equal to 3.83 days) is the parent of all radionuclides in the studied chain. The activity of ^{214}Bi is observed in two separate cases: first in absence and then in presence of ^{222}Rn. The aim is to find the experimental decay constant of ^{214}Bi in both cases. Results allow to verify in which case there is radioactive equilibrium. To measure the activity of ^{214}Bi, a sample containing the radionuclides to be studied was placed in front of an inorganic scintillation detector inside a lead "castle" (to shield from environmental radioactivity). In this way, the detector reveals the gamma rays emitted by the sample due to the

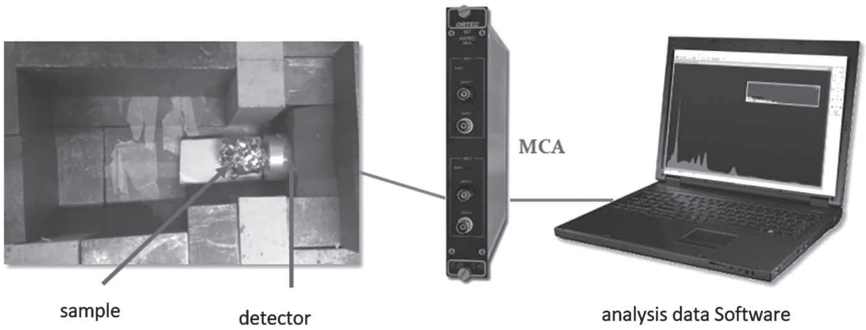

Fig. 3. – Experimental setup.

decay of ^{214}Bi into ^{214}Po; the energy of these photons is 609 keV. The detector signals were analysed through a module (MCA) which provides the energy spectra of gamma rays. A software permitted to view these spectra on a computer screen and to compile a script which automatically acquired them saving a report at each measurement (fig. 3) These reports contained the *net rate* of photons corresponding to the energy of 609 keV. Students recorded this *net rate* as a function of acquisition time because it corresponds to the activity of ^{214}Bi([1]).

4˙1. *The creation of the samples and the measurements.* – To measure the activity of ^{214}Bi first in the absence and then in the presence of ^{222}Rn, it was necessary to design two different methods to prepare two different samples. Both methods required the use of a uranium rock([2]) (fig. 4): samples had to be put in contact with the rock inside a box for three or four days.

4˙1.1. Measurement in absence of ^{222}Rn. In this measurement ^{214}Bi was produced from the decay chain which has as progenitor ^{218}Po, the first decay product of ^{222}Rn (eq. (1)).

$$^{218}_{84}\mathrm{Po}\xrightarrow[t_{\frac{1}{2}}=3{,}03\ \mathrm{min}]{}\,^{214}_{82}\mathrm{Pb}\xrightarrow[t_{\frac{1}{2}}=26{,}8\ \mathrm{min}]{}\,^{214}_{83}\mathrm{Bi}\xrightarrow[t_{\frac{1}{2}}=19{,}7\ \mathrm{min}]{}\,^{214}_{84}\mathrm{Po} \qquad (1)$$

To prepare this sample, in order to have only Radon daughters, the uranium rock was wrapped by aluminium foil inside the box. This allowed to select only Radon daughters on the aluminium foil([3]). Then the foil was taken from the box and placed immediately in front of the detector. Using the automatic script students set an acquisition time

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([1]) The net rate does not correspond exactly to the activity, because it is necessary to take into account some factors which underestimate the real activity: the branching ratio of the decay of ^{214}Bi, and the geometrical acceptance and detector efficiency. However, the proportionality factor is not important for the purpose of the activity trend. For this reason, these factors were not taken into account working with students.

([2]) The uranium rock was collected in Novazza (BG, Italy) where there is a uranium abandoned mine.

([3]) In fact, we suppose that the contamination of the aluminium foil takes place in this way: the Radon, produced by the decay of Uranium, is a gas so it diffuses into the box; when then it decays into its daughter (^{218}Po) this one drops on the aluminium foil and remains attached. Then ^{218}Po continues to decay into the other radionuclides.

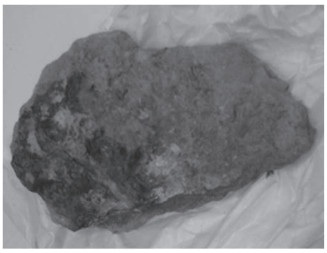

Fig. 4. – Uranium rock.

Fig. 5. – Students work at the measurement.

of 2.5 minutes and a break between two consecutive measures of 30 seconds (fig. 5). Students realized a graph (shown in fig. 6) of *net rate* as a function of time. The errors were calculated directly by the software.

Students using an exponential fit function found the following relation:

$$y = 934.4e^{-0.013x}.$$

The numerical coefficient of the exponent was in good agreement —same order of magnitude— with the theoretical value of the constant decay of ^{214}Bi as shown in table I. This implies that a radioactive equilibrium is not established within this radioactive family.([4])

([4]) A rigorous data analysis would require to fit the data with a numerical simulation using the Bateman equations. In this way, it would be possible to take into account the decay of all radionuclides within the family (^{218}Po and ^{214}Pb). For reasons of time students used a simple exponential function. This involved obviously that the experimental results were not in excellent agreement.

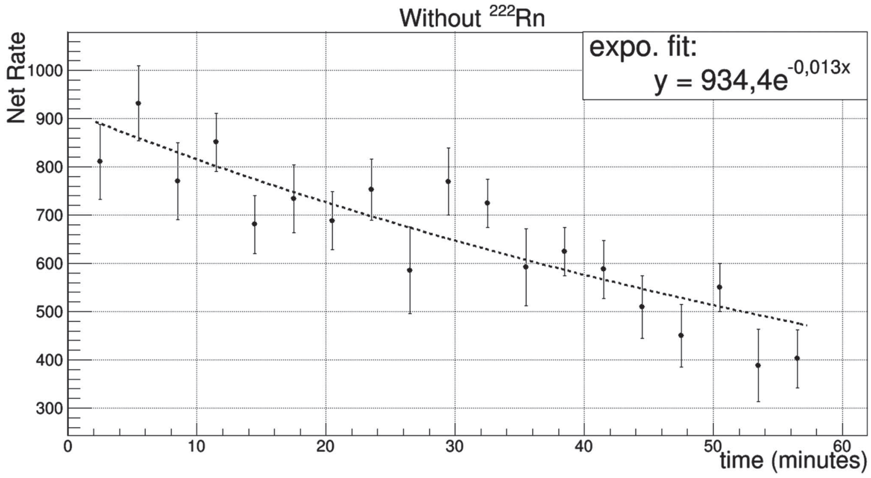

Fig. 6. – Activity trend of ^{214}Bi as a function of time in the absence of ^{222}Rn.

TABLE I. – *Comparison between the experimental value obtained from the fit and the theoretical values of the decay constants of* ^{214}Bi *and* ^{218}Po.

Theo λ^{218}Po (min^{-1})	Theo λ^{214}Bi (min^{-1})	Exp λ (min^{-1})
0.229	0.034	0.013

4˙1.2. Measurement in presence of ^{222}Rn. In the second measurement, ^{214}Bi was produced by the following decay chain (eq. (2)).

$$^{222}_{86}\text{Rn} \xrightarrow[t_{\frac{1}{2}}=3{,}83\ \text{days}]{} {}^{218}_{84}\text{Po} \xrightarrow[t_{\frac{1}{2}}=3{,}03\ \text{min}]{} {}^{214}_{82}\text{Pb} \xrightarrow[t_{\frac{1}{2}}=26{,}8\ \text{min}]{} {}^{214}_{83}\text{Bi} \xrightarrow[t_{\frac{1}{2}}=19{,}7\ \text{min}]{} {}^{214}_{84}\text{Po} \tag{2}$$

To prepare the sample so that even ^{222}Rn could be included, it was necessary to trap the Radon. Among the possible techniques used to trap radon [7-9], the one used in this experience was very simple and consisted in making a solution of radon in ethyl alcohol. Ethyl alcohol was chosen because the solubilty coefficient of radon in alcohol is about six times greater than that one of radon in air. To make the solution, an alcohol filled ampoule was kept open in the box with the uranium rock. In this way radon, spreading out in the box, dissolved in the alcohol. Then, the ampoule was taken from the box, corked and placed in front of the detector (fig. 7).

Students, using the automatic program, set a 30 minutes acquisition time. Subsequently, they collected all data from the reports corresponding to a week of acquisition. The errors were calculated directly by the software. The graph of *net rate* as a function of time is shown in fig. 8.

Using an exponential fit, students obtained this relation

$$y = 3362e^{-0.000127x}$$

The numerical coefficient of the exponent ($12,71\cdot10^{-5}$ min^{-1}) was completely different from the value obtained previously (there was a difference of three orders of magnitude)

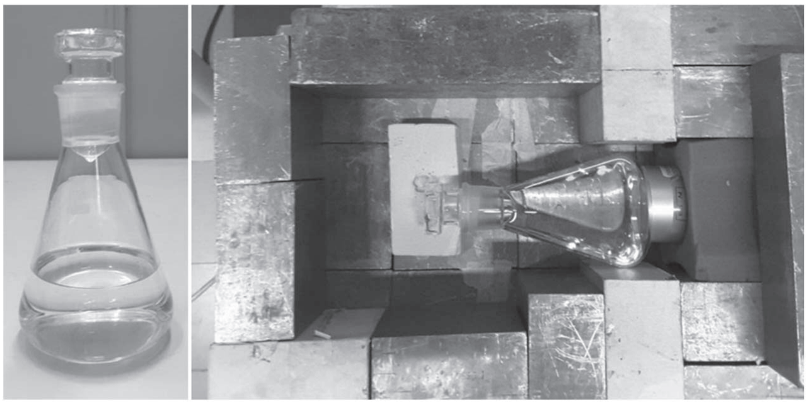

Fig. 7. – On the left, the ampoule with ethyl alcohol inside. On the right, the ampoule in front of the detector before starting the measurement.

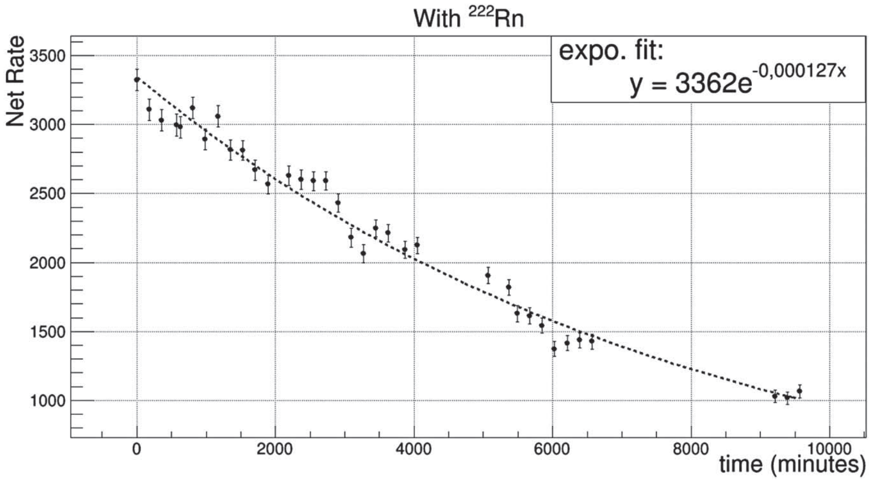

Fig. 8. – Graph of the activity of ^{214}Bi as a function of time in the presence of ^{222}Rn.

but indeed it was in good agreement (within 1%) with the theoretical value of ^{222}Rn ($12,57 \cdot 10^{-5}\,\mathrm{min}^{-1}$). Therefore, the decay constant of ^{214}Bi was quite similar to the decay constant of ^{222}Rn. Observing the activity of ^{214}Bi, students measured the decay constant of its progenitor: so, radioactive equilibrium between the two radionuclides was established. This was possible because the decay constant of ^{222}Rn is much lower (about 270 times) than that of ^{214}Bi. To complete the experience, students developed a numerical simulation of the decay of a radioactive family composed of only two radionuclides: a parent and a daughter. In this simulation, realized through an Excel worksheet, students used the mathematical equations of radioactive decay to reproduce the trend of both activities as a function of time. The free parameters of the simulation were the initial number of radionuclides and the two decay constants. Including these parameters in the decay equations, students calculated the number of radionuclides lost in time interval $\mathrm{d}t$ provided that $\mathrm{d}t \ll \tau_1$ and $\mathrm{d}t \ll \tau_2$ where τ_1 and τ_2 are the mean life times of the two radionuclides. Finally, students calculated the number of remaining radionuclides. The first parameters that students used to test their simulation were the decay constants of ^{214}Bi and ^{222}Rn to check that an equilibrium was really established between them. Students observed the trend of both activities as a function of time and checked numerically that the ratio of activities tended to a constant value: in the case of Radon and ^{214}Bi the constant value was approximately 1.003. This led to the conclusion that it was possible to confirm the establishment of a secular equilibrium (within a 0.3% difference). As further evidence, students used the decay constants of ^{218}Po and ^{214}Bi and found that equlibrium was not established. Varying the decay constants, they were able to create different equilibrium situations and to understand what kind of equilibrium (transient or secular) was formed simply calculating the ratio between the activities of the two isotopes. This numerical simulation allowed students to strengthen the concept of radioactive equilibrium and the difference between transient and secular equilibrium.

5. – Conclusion and future developments

Through this experience, students have the opportunity to approach radioactivity different practices:

- The experimental observation of the decay of some radionuclides belonging to the same radioactive chain and the estimate of their decay constant to determine whether radioactive equilibrium is established.
- The use of natural radioactive samples and of instruments commonly used in nuclear spectroscopy experiments and techniques. Such techniques are adopted to set up experimental conditions suitable for the detection of gamma rays, in compliance with all safety requirements.
- The creation of a model as a valuable tool to understand and predict a physical phenomenon. It is important to know the degree of approximation of the model in order to understand its limits. The dice model allows to highlight how the exponential mathematical model can represent different phenomena governed by a constant probability to "decay" (changing state irreversibly from one state to another). A simple random process (rolling dice and discarding them according to a certain rule) can be used to model a physical phenomenon governed by the same rules. This becomes very useful when one wishes to investigate what happens when these phenomena are combined in more complex way (*e.g.* "in series" as in the case of radioactive chain).
- The numerical simulation by using the mathematical relations of radioactive decay. Such simulation allowed to compare with theoretical predictions both the experimental data and the results of the dice model.

Students were generally satisfied with this activity and this encourages us to plan other experiences for high school students in the future.

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The activity presented was designed and performed as a collaboration between P. Vitulo (Department of Physics, University of Pavia and INFN Pavia), P. Montagna (Department of Physics, University of Pavia and INFN Pavia), A. De Ambrosis (Department of Physics, University of Pavia), M. Malgieri (Department of Physics, University of Pavia) and the author of the present article, who coordinated the collaboration.

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