

# Experimental Search for Molecular-Nuclear Transitions in Water\*

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Experimental search for molecular-nuclear transitions  $\text{H}_2\text{O} \rightarrow {}^{18}\text{Ne}^*(4.522, 1^-) \rightarrow {}^{18}\text{F} \rightarrow {}^{18}\text{O}$  in water molecules was carried out. The measurements were performed in a low-background laboratory at the Baksan Neutrino Observatory. Under the assumption that the above transitions take place, the estimate for the half-life time of water molecule was found to be about  $10^{18}$  years.

## I. INTRODUCTION

Phenomenon of a long-range effective interaction in some quantum systems including constituents which are only interacting at short distances is known already for a long time. Here, by a short-range interaction we understand any interaction that falls off at large distances between particles, i. e. as  $r \rightarrow \infty$ , not slower than exponentially. In the context below, long-range forces decrease at infinity as some inverse powers of  $r$ .

The most pronounced manifestation of this phenomenon appears in the so-called “Efimov effect” [1]. The Efimov effect consists, in particular, in arising an effective interaction between a particle and a coupled pair of particles which behaves as  $1/r^2$ . A sufficient condition for this effect to take place in a three-bosonic system is a closeness of the binding energies at least for two of pair subsystems to zero. Just the very small binding energies force the two-body wave function to be extremely extended generating an effective long-range interaction of the coupled pair with a complementary particle.

Another example of a long-range effect of short-range interaction was found by Ya. B. Zeldovich [2]. He showed that if two particles interact via two potentials, a short-range one and an arbitrary long-range (say, Coulomb) one, then the spectrum of the two-body system can be drastically changed as compared to the case of a long-range potential only. This change takes place if the short-range potential generates a bound or resonant state with an energy sufficiently close to zero.

Thus, in both the above cases, long-range effects take place due to the fact that a short-range interaction causes a two-body system to be, nevertheless, very extended. Having this in mind, one can expect some enhancement for the probability of transition of the system

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TABLE I. Two examples of nuclear systems with near-threshold resonances

Molecular System	Few-Atomic System		Composed Nuclear System (CNS)			
	Nuclear Subsystem (NS)	Energy of NS over g.s. of CNS (MeV)	Nucleus	$J^\pi$	E(MeV)	$\Gamma(\text{keV})$
$\text{H}_2\text{O}$	$^{16}\text{O}+p+p$	4.522	$^{18}\text{Ne}$	$1^-$	4.519	$< 9$
$\text{OH}^-$	$^{17}\text{O}+p$	5.607	$^{18}\text{F}$	$1^-$	5.604	$< 1.2$

from a molecular state to an extended (resonance) nuclear state as compared to a similar transition into a localized nuclear state. Indeed, model calculations [3] of the overlap integral between wave functions of the  $\text{H}_2\text{O}$  ( $1^-$ ) molecule and a resonant state (4.522,  $1^-$ ) of the nucleus  $^{18}\text{Ne}$  show an enhancement of this sort.

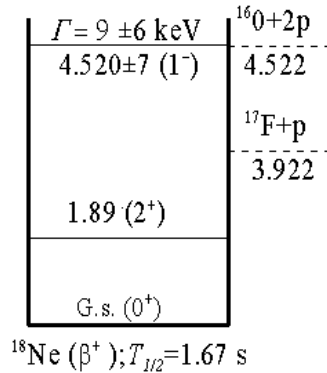
The purpose of the present experiment is to estimate the life time of water molecules with respect to the following decay chain

$$[\text{H}_2\text{O}]_{1^-} \longrightarrow ^{18}\text{Ne}^* (4.522, 1^-) \longrightarrow ^{18}\text{Ne} (\text{g. s.}) \longrightarrow ^{18}\text{F} \longrightarrow ^{18}\text{O} + \beta^+.$$

## II. EXPERIMENTAL APPROACH AND RESULTS

A number of examples of nuclear systems with near-threshold resonances were analyzed from this point of view, among them are (p, p,  $^{16}\text{O}$ ) and (p,  $^{17}\text{O}$ ) [4,5], i.e. the nuclear constituents of the usual water molecule  $\text{H}_2\text{O}$  and hydroxyl ions  $\text{OH}$  based on the rare oxygen isotope  $^{17}\text{O}$  (see Table I).

For the first experimental study in this direction we choose the system (p, p,  $^{16}\text{O}$ ), i.e. the  $\text{H}_2\text{O}$  molecule. Its properties, in addition to obvious availability, make this case the most favorable for the experiment. From Fig. 1, displaying the energy-level diagram for  $^{18}\text{Ne}$ -nucleus, it is seen that rotation states  $1^-$  of a water molecule and a highly excited state ( $1^-, 4.522$  MeV) of this nucleus can be considered as energy degenerate. Thus, a real physical state with these quantum numbers appears to be a superposition of molecular and nuclear states. Experimental approach was designed with taking into account of the properties of both the components of the superposition pair.

FIG. 1. Fragment of the nucleus  $^{18}\text{Ne}$  spectrum.

As a rule, rotational states are excited only in free molecules while their population under the conditions of condensed-phase water is prohibited due to powerful hydrogen bonds [6]. This makes carrying out the experiments more difficult, but at the same time it gives additional opportunities to manipulate with the hypothetical process of molecular-nuclear transitions in a water system. So, it becomes possible to use an accumulation-measuring cycling during searching for resulting radioactive products. The layout of measurements is shown in Fig. 2. The accumulation cycle represented heating the water portion within a sealed out measuring chamber to a critical point about 647 K, at which a total amount of water was for sure in a vapor phase regardless of pressure within the volume. To withstand this rather a high pressure (22.5 MPa [7]), the stainless-steel or titanium chambers were strengthened for the heating period by two thick steel removable plates at the top and bottom of the chambers. The latter were thick-wall shortened metallic cylinders, sealed at the faces by thin membranes almost fully transparent for the expected annihilation radiation  $E_\gamma = 511$  keV.

For the measuring period, the plates were removed, and the chamber was placed after cooling between two NaI(Tl)-scintillators, operating in the  $\gamma\gamma$ -coincidence mode. First, the measurements were performed in surface-laboratory conditions at the Institute of Physics and Technology Problems (Dubna), then the main part of the experiments was undertaken, this time at a deep underground laboratory of the Baksan Neutrino Observatory (Republic of Kabardino-Balkaria, the North Caucasus) of the Institute for Nuclear Research of the Russian Academy of Sciences. At the surface-laboratory stage, necessary measurement procedures were optimized, and a yield of molecular-nuclear transition in water in the condensed state was estimated. Analysis of these data showed main sources of the background counting: true coincidences due to the cosmic muons, decay of the natural  $^{40}\text{K}$  and daughter products of  $^{222}\text{Rn}$  ( $^{214}\text{Bi}$  and  $^{214}\text{Po}$ ). To take into account the background in the region of “interest”, i. e., near  $E_\gamma = 511$  keV, some control background measurements were carried out under the identical conditions (geometry, amount of water, *etc.*), in which heavy water ( $\text{D}_2\text{O}$ , 99.0%-enrichment) was used instead of the natural one. Within the limits of statistical fluctuations, both the spectra were identical. For the water half-life in the condensed-phase state with respect to decay by the chain  $\text{H}_2\text{O} \longrightarrow {}^{18}\text{Ne}(\beta^+; 1.7\text{ s}) \longrightarrow {}^{18}\text{F}(\beta^+; 109\text{ min})$ , a lower limit was estimated as  $T_{1/2} \geq 4.10^{21}$  years (within the 99%-confidence level). At the Baksan Observatory, all the efforts were made to subdue the background radiation as much as possible. The cosmic muons were subdued due to power shielding, as the measuring premises were situated inside a gallery created within the mountain Andyrchi (one of mountains of the Elbrus environment, where the screening thickness of a rock achieves about 600 m of water equivalent. Walls (the ceiling and floor, as well) were built of a special uranium-free concrete. The scintillators were prepared on the basis of materials with a low containment of potassium and radium. They were, for a long time, located in a deep underground. Thus all short-lived cosmic-ray-induced activities should be extinct. The detection unit was provided with an additional shielding composed of interchanged layers of pure tungsten, lead, and copper.

For every  $\gamma$ -quantum event, the time was registered with a channel width 10 s. This was more than sufficient for a detailed analysis of the time dynamics in the range of interest for this experiment. The accumulation interval was chosen to be about three half-lives of  $^{18}\text{F}$ , i. e. 5.5 hours. The measuring time-schedule for these runs is inserted in Fig. 3. Intervals of about four half-lives of  $^{18}\text{F}$ , i. e.  $\sim 440$  min each, were considered as time-periods when the decay of hypothetically accumulated  $^{18}\text{F}$  could yet give some contribution to the counting in the region of  $E_\gamma = 511$  keV. The remaining part of measuring cycles was used to calculate

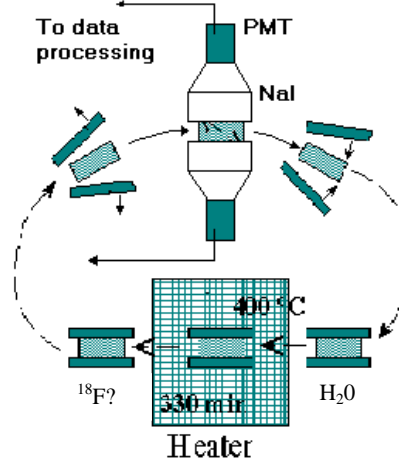


FIG. 2. Scheme of accumulation-measuring procedure.

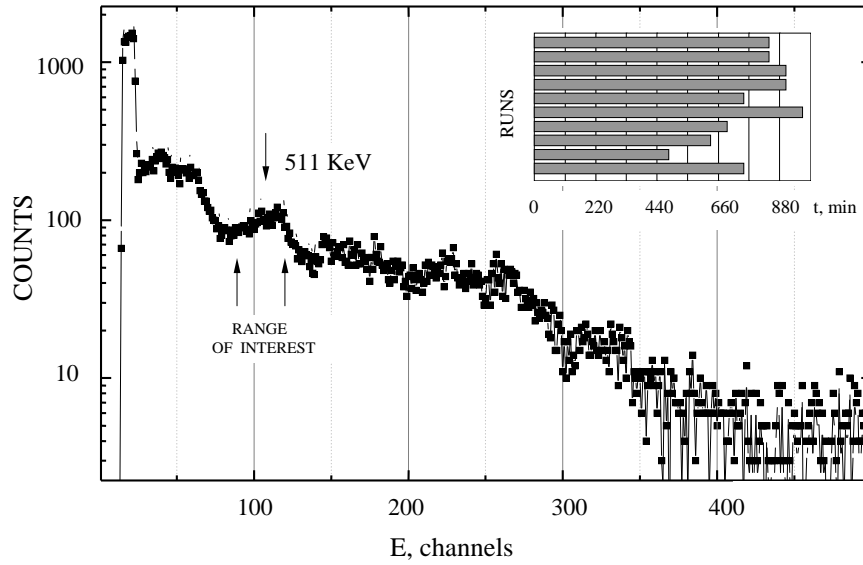


FIG. 3. The  $\gamma\gamma$ -coincidence spectrum obtained during ten measuring cycles. Marked by arrows is an energy region around  $E_\gamma = 511$  keV. Time-durations of measuring periods are displayed in the insertion to the graph.

the background counting in the same energy range.

Preliminary analysis of the results was carried out by means of comparing counting rates in two above measurement periods, each of a total duration  $\sim 4000$  min for ten cycles. A certain excess (at a level about 1-2 RMS deviation) of the counting rate in the region of  $E_\gamma = 511$  keV was observed in the first period as compared to the second one, i.e. to the background. We emphasize that the above excess was non-stationary. Time dependence of the non-stationary process approximately corresponded to the half-life of  $^{18}\text{F}$ . Under the assumption that the observed non-stationary component of the effect is associated with the accumulation and decay of the nuclei  $^{18}\text{F}$ , our estimate for the efficient half-life of the water molecule with respect to the nuclear channel under consideration is  $T_{1/2} \sim 10^{18}$  years.

Although the total statistics was not sufficient to make a decisive conclusion, the results obtained are rather encouraging for further experiments in this intriguing direction with new more sensitive approaches. More attention should be paid to analysis of population rates of suitable molecular states under the experimental conditions. Also some further theoretical consideration are desirable regarding the fusion probability estimates for this and other molecular systems.

## ACKNOWLEDGMENTS

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