1.1 **Investigation of the channels of excitation and decay of the anomalously low-energy isomeric level 3/2+ (3.5±1.0 eV) in the Th-229 nucleus**.

1.2 1.100

1.3 24

1.4 July-96

1.5 5

1.6 60.000

1.7 It is confirmed by the signatures put below, that the information contained in the present application is true and exactly corresponds to the information, obtained from all participants of project. All the project participants took part in the preparation of this application and agree with it.

The application contains 9 pages.

Project Coordinator

Romain Coussement

December 8, 1995

Russian Principle Investigator

Tkalya, Eugenii Viktorovich

December 8, 1995

2.1 CO
2.2 1
2.3 Professor
2.4 Director
2.5 M
2.6 Romain
2.7
2.8 Coussement
2.9.1 Instituut voor Kern-en Strahlingsfysika, Katholieke Universiteit Leuven
2.9.2
2.9.3
2.10
2.11 Celesrijnenlaan 200 D
2.12
2.13 B-3001
2.14 Leuven
2.15 BE
2.16 +32-16-327260
2.17 +32-16-327985
2.18 romain.coussement@iks.kuleuven.ac.be
2.19 2
2.20 9,000
2.21 0
2.22 0
2.23 4,000
2.24 0
2.25 5,000
2.26 0

2.1 PI
2.2 2
2.3 Doctor of physical and mathematical sciences
2.4 Head of laboratory
2.5 M
2.6 Eugenii
2.7 Victorovich
2.8 Tkalya
2.9.1 Nuclear Safety Institute
2.9.2 Department of Radiation Safety
2.9.3 Laboratory of Theory of Radiation Processes
2.10 RAS
2.11 Bolshaya Tylskaya 52
2.12
2.13 113191
2.14 Moscow
2.15 RU
2.16 +7-095-9552888
2.17 +7-095-2302029
2.18 pbl@ibrae.msk.su, Subject: Tkalya
2.19 2
2.20 16,000
2.21 8,000
2.22 0
2.23 8,000
2.24 0
2.25 0
2.26 0

2.1 CR
2.2 3
2.3 Professor
2.4 Senior Scientist
2.5 M
2.6 Giovanni
2.7
2.8 Principi
2.9.1 Universita Di Padova
2.9.2 Dipartimento Di Ingegneria Meccanica
2.9.3 Settore Materiali
2.10
2.11 Via Marzolo 9
2.12
2.13 35131
2.14 Padova
2.15 IT
2.16 +39-49-8275513
2.17 +39-49-8275510
2.18 PRINCIPI@IPDUNIVX.UNIPD.IT
2.19 2
2.20 9,000
2.21 0
2.22 0
2.23 4,000
2.24 0
2.25 5,000
2.26 0

2.1 CR
2.2 4
2.3 Doctor of physical and mathematical sciences
2.4 Professor
2.5 M
2.6 Alexandr
2.7 Mikhailovich
2.8 Dykhne
2.9.1 Moscow Institute of Physics and Technology
2.9.2 Department of physics problems of energetic
2.9.3 Chair of applied physics
2.10 GosCom-Vuz
2.11 Institytskii pereylok 9
2.12
2.13 141700
2.14 Dolgoprudnyi, Moscow reg.
2.15 RU
2.16 +7-095-3332155
2.17 +7-095-3332155
2.18
2.19 2
2.20 16,000
2.21 8,000
2.22 0
2.23 6,000
2.24 2,000
2.25 0
2.26 0

2.1 CR
2.2 4
2.3 Candidat of physical and mathematical sciences
2.4 Head of laboratory
2.5 M
2.6 Valerii
2.7 Victorovich
2.8 Lomonosov
2.9.1 Institute of General Nuclear Physics, Russian Research Center the "Kurchatov Institute"
2.9.2 Department of Nuclear Physics and Radiochemistry
2.9.3 Laboratory of Collective Interactions
2.10
2.11 Kurchatov Sq. 1
2.12
2.13 123182
2.14 Moscow
2.15 RU
2.16 +7-095-1969339
2.17 +7-095-8825804
2.18 vvl@iae.msk.su
2.19 2
2.20 16,000
2.21 8,000
2.22 0
2.23 6,000
2.24 0
2.25 2,000
2.26 0

3.1 The title of project.

Investigation of the channels of excitation and decay of the anomalously low-energy isomeric level 3/2+ (3.5±1.0 eV) in the Th-229 nucleus.

3.2 Project objectives and state-of-the-art review.

The main objectives of the Project is a comprehensive study of the anomalously low-energy isomer level in the Th-229 nucleus of some unique phenomena and processes, non-studied so far, which are closely connected with the existence of the above mentioned isomer.

Among the most important ones are the following

1. Excitation of the low-energy 3.5±1.0 eV nuclear level in Th-229 by surface plasmons and laser radiation including the process of excitation via the atomic shell by the inverse atomic bridge mechanism.
2. Study of the channel of the low-energy isomeric state decay: direct nuclear radiation in the optical region and the electron bridge, third-order process. A refinement of the energy and determination of the level halflife.
3. Development of a new precision method for measuring fine effects in the electron-nucleus and phonon-nucleus interaction and the effect of chemical environment on atoms, basing on the results obtained.
4. Generation and studying of the coherent gamma-radiation in the optical range. Development of a "nuclear"source of light with reference narrow line.
5. Moessbouer spectroscopy in the optical range.

The new experimental data obtained at the National Engineering laboratory in Idaho (USA) in 1989-1993 evidence definitely that in the Th-229 nucleus the 3/2+ level, being the base of the 3/2[631] rotation band, has the optical energy 3.5±1.0 eV. However the isomeric nuclear M1 transition between this level and the ground state 5/2+ was not observed because of the weak population of the isomeric level in the gamma transition from the high-energy sates of Th-229, populating in the alpha-transition of U-239. Therefore, neither the lifetime of the low energy isomer nor its decay channels have been determined so far.

In the Project a quite different approach to studying the whole set of problems associated with the anomalously low-energy level in Th-229 is proposed. For the realization of the program presented the excitation of a grate number of Th-229 nuclei by the laser radiation and surface plasmons by the inverse electron bridge mechanism will be first accomplished. This process of the nuclear excitation not "directly" but through the atomic shell, serving a certain electron bridge between the incident radiation and the nucleus, can provide a high efficiency of the Th-229m nuclear excitation inspite of great range of uncertainty - 2 eV per isomeric level energy magnitude.

At present the project proposed has no analogies by a large number of parameters. For nuclear physics the most important process seems to be observation of the isomer decay through the electron bridge, the third-order process, whose existence has not been experimentally provided so far, and the determination of the properties of the M1-transition between two rotation bands, forbidden by asymptotic quantum numbers. Of great interest for optics and laser physics is the process of excitation of the nuclear level by the laser radiation and creation of the inverse population. Here serious perspectives open. In particular, it is sufficient to mention the nuclear transition level in the optical range and the nuclear source of light with a reference-narrow line. For the surface physics studies of the nuclear interaction with plasmons (electro-magnetic waves propagation in vacuum at the boundary with the surface-active media are undoubtedly important. Of interest for solid state physics is the Moessbouer spectroscopy in the optical range and the possibility to use the Th-229M isomer as an extremely sensitive probe for studying the chemical environment, phonon and electron nuclear interactions and other fine effects.

All the above features make the project interesting for physics as a whole. The theoretical calculations made for five recent years show that the above problems are complicated but solvable. For this purpose the following Institutions and teams should be invited to participate in the work.

From the Russian side:

1. Moscow Institute of Physics and Technology. The team headed by Professor A.M. Dykhne, Academician of the Russian Academy of Sciences. This team is carrying out theoretical and experimental studies on interactions between the laser radiation and the substance, participates in the works on the excitation of the Th-229 isomer by surface plasmons. They have up-to-date experimental basis in their disposal.

2. Nuclear Safety Institute the Russian Academy of Sciences (IBRAE RAN). The team headed by E.V. Tkalya, Dr. Sci.(Phys.Math.) For last five years this team has carried out the theoretical studies forming the

basis of the present project: the decay channels have been determined depending on the isomer energy; estimation of the life times have been made; cross-sections of the resonant excitation of the Th-229 nuclear level by optical phonons and surface plasmons have been calculated. At present this team is participating in the experimental studies on Thorium isomer excitation by laser radiation.

3. Institute for general Physics of the Russian Research Center "Kurchatov Institute". The laboratory headed by V.V. Lomonosov, Cand. Sci.(Phys.Math). At present the team is carrying out experiments on Thorium isomer excitation by laser radiation. They have an extensive experience with theoretical and experimental studies of surface plasmons and Moessbouer radiation properties. The team has all necessary equipment for works with radioactive substance, including radiochemical studies.

From the INTAS member states:

1. Institute of Nuclear and Radiation Physics of Leuven Catholic University (Belgium). The team headed by Professor R. Coussement. This Institute has a new unique experimental equipment for production and studies of the properties of radioactive elements. It is supposed to carry out here a part of experimental studies on the excitation of the Th-229 isomer by surface plasmons.

2. Padova University (Italy), Faculty of Engineering Mechanic, Sector of Materials. The team headed by Professor G. Princippi. This group is engaged in studying the properties of surface and Solids by nuclear physics methods, has the experience of and equipment for preparation of high quality targets with Th-229 on thin films and metallic substrata would be prepared.

3.3. Scientific and technical description.

3.3.1. Trends of studies.

The theoretical calculations and preliminary experiments show that it is not possible to determine the unknown characteristics of the low-energy level of the Th-229 nucleus such as energy, halflife, decay channels etc, using the conventional nuclear spectroscopy methods (measurements of the Th229 gamma spectrum accompanying the U-233 alpha decay): the resolving power of the detection instrumentation is too low, the capability of the statistical analysis of the results of many-year spectrometric measurements have been exhausted.

In this situation another approach seems to be the most suitable, e.i. the excitation of a great number of Th-229 isomeric nuclei from the ground state with the subsequent measurement of the isomeric level decay characteristics at high signal/noise ratios. The fact that the isomer's energy is in the optical range, from 2.5 eV to 4.5 eV, allows the laser with a suitable wave length or surface plasmons of the desired energy to be used for this purpose. (The surface plasmons are the electromagnetic waves propagation on the boundary between different media,e.g. the vacuum/conductor interface.

The great uncertainty in the isomer energy (2 eV is a very high value for the optical range) makes senseless the attempts to get the direct nuclear photoexcitation in the first experiments. It will take several years to pass through the existing uncertainty range with the narrow laser line irradiating the target for at least 100sin each step. Therefore we propose here an alternative scheme: to excite the nuclear isomer by photons or plasmons through the atomic shell serving as an electron bridge. As shown by the calculations the presence of a significant (several hundreds) number of excited nuclear states with relatively large (as compared with the nuclear ones) radiation widths should essentially facilitate the excitation process. In this case the atom affected by the radiation of the tunable laser transforms virtually into the specified excited state which then transfers a part of the excitation energy from the atomic shell to the nucleus in one of its partial decay channels. The process was called "the inverse electron bridge" (IEB). Due to very low energies the cross-section of this third order process may become much larger, under certain conditions, that of the first-order process, the direct excitation (it should be noted that this is a rather typical in nuclear spectroscopy). Therefore it is far easier to realize the resonant excitation of the nucleus via a system of excited atomic states than "directly". These are the properties of the electron bridge that will be used in the first experiments. The theoretical calculations show that even at a relatively large detuning of the order of 0.1 eV in the energies of the nuclear and the "second" atomic transitions (it does not seem reasonable to take a higher value taking into account the high density of the excited states in the Thorium atom) the isomer's resonant excitation cross-section is about 10^{-21}—10^{-22} cm^2. Correspondingly, in irradiation of a 1 mm diameter target containing 10^{-6} g of Th-229 (about $3{\cdot}10^{15}$ nuclei) the 100 mW laser with a conventional line width 10^{-5}—10-6 eV will "produce" about 10^{12} isomers per second. At such an excitation rate sufficient quantities of the isomer nuclei will be produced for relatively short times to ensure reliable observation of the decay process even at long lifetimes. Of the low-energy isomers.

The scheme of the "laser" experiment is as follows. Thorium atoms with the Th-229 isotope, implanted into the a transparent matrix, are irradiated with the laser which is alternatively tuned to the well known wavelengths of the Thorium atomic transitions. In adjustment to one of these transitions the resonant excitation of Thorium isomers begins, of course, with a much smaller cross section than that of the resonant nuclear excitation. (Here will affect the smallness introduced by the factor, the squared ratio of the laser line width to the above mentioned value of the difference between the energies of the nuclei and second atomic transitions, which always appears in such cases.) In the most unfavourable case the cross section will reduce by 8—9 orders of magnitude, and minimum excitation rate will be 10^3 — 10^4 nuclei/sec. Upon detection of a slight signal indicating the isomer decay, it is necessary to perform scanning around this line, going step-by-step away from the atomic transition. During a few "steps" this will give an increase in the nuclear excitation rate. The peak of the excitation rate will correspond to the resonance tuning of the laser radiation to the nuclear transition through the system of atomic levels. This will permit the isomer's energy to be rapidly determined by the known energies of atomic states and laser photons.

The experiment with surface plasmons is somewhat simpler. There are effective methods for the surface plasmon generation on the metal/vacuum interface using the electron beam or the laser radiation with possible generation of plasmons including those of the visible and ultraviolet ranges. In this energy region, when using the laser beam, either the method of disturber total inner reflection in Kretschmann geometry, or the scheme of lattice conversion of the laser radiation into the surface electromagnetic wave are usually used. However, taking into account the nuclear transition energy uncertainty scale as well as the available instrumentation and experimental equipment, it is more reasonable to use the electron beam in the first experiments. The desired spectrum of surface plasmons with the common envelope with the width of 1—2 eV around the central frequency 3.5 eV is then easily obtained using the suitably formed electron beam with the required electron energy scattering. The calculation shows that the excitation process via the IEB mechanism has here a cross section of the order of 10^{-25} — 10^{-26} cm^2. As the plasmons with energies 3—4 eV have the path length of about 10^{-3} cm and propagate in vacuum (air) in the 10^{-5} cm thick near-surface layer, the effective density of the surface plasmon flow proves to be two orders of magnitude higher that the density of the beam of the target-irradiating electrons. Thus, for target containing the same number of $3{\cdot}10^{15}$ Th-229 nuclei, deposited on the silver or aluminum substratum, at the 100 mA electron current the excitation rate will be higher than 10^{11} nuclei/sec.

Depending on the level energy the most probable channels of the Th-229 isomer decay are either the direct nuclear radiation in the optical range with the lifetimes in hours and longer or the electron bridge with the probable lifetime in seconds or longer. Within the range from 2.5 eV to 4.5 eV the expected lifetime in the neutral Thorium atom is somewhere between 10 s and 2 days. Taking into account this preliminary values the irradiation of the order 100 s should be taken. This is sufficient for the activity of decaying isomer Thorium nuclei exceeded 10^7 — 10^8 Bq. The optical photons obtained from the direct nuclear radiation and a pair of photons through the electron bridge should be detected. The sum of the energies of two "soft" photons from the electron bridge is always equal to the energy of photons from the direct nuclear radiation.

It is supposed to go away from the background created by alpha-particles bremsstrahlung in the target material (Th-229 is alpha-active with the halflife of 7880 years), using the standard coincidence methods tested on the Thorium targets. For the targets with the surface Thorium-229 nucleus arrangement the photon alpha-coincidence method is used, for a target made of a transparent matrix with implanted Thorium nuclei, the photon-gamma coincidence method is used (the alpha decay of Thorium-229 is accompanied with gamma irradiation and characteristic X-radiation.

The above method for obtaining a large quantity of isomers allows Moessbouer spectroscopy methods to be used for studies of the substance structure. It is supposed to eliminate the characteristic radiation line broadening resulting from the nucleus and atom interaction with the chemical environment using one of the standard methods: in-freezing of the Thorium atoms into the inert gas matrix, injecting them to the laser trap or by "dissolving" them in liquid Helium-4.

The inverse population created in the system of Th-229 isomeric nuclei also permits the process of the nuclear coherent radiation to be studied. The electron bridge in Thorium passes through the excited state of the valence electrons of the atom. Therefore, changing the chemical compounds of the Thorium atom with the Th-229m isomeric nuclei, the partial decay width can be easily affected through the electron bridge, including also the change towards its reduction (for example, in the metal where a part of valence electrons is generalized, the probability of the decay via the electron bridge will be close to zero). Succeeding in the reduction in this width to the values comparable or lower than the partial width of gamma radiation, one may try to reach the increase in the radiation in a sample of specific geometrical shape.

This project based on the scientific results, published in the following papers.

1. E.V.Tkalya. The Cross Section of Excitation of the Low-Lying (<5 eV) Isomer of the Nucleus of Th-229 by Laser Radiation via the Inverse Electronic Bridge Mechanism. // Sov.J.Nucl.Phys. 55 (1992) 1611.
2. E.V.Tkalya. Excitation of the Low-Lying Isomeric Level in the Th-229 Nucleus by Optical Photons. // JETP Lett. 55 (1992) 211.
3. V.F.Strizhov and E.V.Tkalya. Decay Channels of the Low-Lying Isomeric State of Nucleus Th-229. The Possibilities of Experimental Researches. // Sov.Phys. JETP 72 (1991) 387.
4. E.V.Tkalya. Nuclear Excitation in Atomic Transitions (NEET Process Analysis). // Nucl.Phys.A539 (1992) 209.
5. V.V.Lomonosov and M.Yu.Talantov. Emission of Radiation by a System of Identical Excited Three-Level Emitters. // JETP 78 (1994) 144.
6. E.V.Tkalya, V.O.Varlamon, V.V.Lomonosov et al. Processes of the Nuclear Isomer Th-229m(3/2+, 3.5±1.0 eV) Resonant Excitation by Optical Photons. // Phys.Scr. 52 (1995) N6.
7. G.Neyens, I.Van Asbroeck and R.Coussement. Static quadrupole moment of high-spin isomers in the doubly-odd 214 Fr nucleus. // Phys.Rev.C51 (1995) 3483.
8. R.Coussement, G.Neyens, M.Van Den Bergh et al. Amplification of gamma-radiation with hidden inversion. // Laser Physics 5 (1995) 292.
9. R.Coussement, G.Neyens, M.Vam Den Bergh et al. Resonant emission without absorption. // Hyperfine Interactions 95 (1995) 13.
10. G.Neyens, R.Nouwen, R.Coussement et al. The static quadrupole moment of high-spin isomers in 210Rn, 211Rn and 214Fr measured with LEMS. // Proc.Int.Conf. on Nuclear Shapes and Nuclear Structure at Low Excitation Energies (ed. M. Vergnes et al) Frontiers p. 221 (1995)
11. G.Neyens, R.Coussement, S.Michiels et al. Quantum Interferences in Moessbauer Transitions? // Proc.Int. Conf. on the Applications of the Moessbauer Effect, p. 12.31. Italy, 1995.
12. G.Neyens and R.Coussement. Quantum Optics With Nuclear Gamma-Radiation. // Proc.Int.Conf. on the Application of the Moessbauer Effect, p.13.1. Italy 1995.

3.3.2. The scientific results. The results of the studies will be presented in the form of articles in scientific journals and submitted at the international conferences

3.4. Management information.

3.4.1. Distribution of tasks.

The team headed by Prof. R. Coussement (Institute of Nuclear and Radiation Physics, Leuven, Belgium) will prepare the experimental equipment and participate in the performance of the experiments on Thorium-229 isomer excitation by surface plasmons. Plasmon will be generated by the electron beam. The team also will study the Moessbouer effect in the optical range in the nuclear transition in Th-229.

The group headed by Prof. G. Principi (Padova University, Italy) will fabricate Thorium targets on thin films and on surface-active media. It also will perform target quality control. The target will be prepared on the basis on an optically transparent matrix.

The team headed by Professor A.M. Dykhne (Moscow Physical and Technical Institute, Russia) will carry out theoretical and experimental studies on excitation of Th-229 isomer by the laser radiation and surface plasmons (plasmons are generated by the laser radiation). The group will engaged in solving the experimental problems on development of a nuclear source of light with the reference line and on production of the nuclear coherent radiation on the basis of Th-229.

The group of E.V. Tkalya, Dr. Sci. (Phys-Math) Nuclear Safety Institute the Russian Academy of Sciences (IBRAE RAN), will provide theoretical studies of the problems of the Thorium isomer excitation by the laser radiation and surface plasmons, carry out studies of the isomeric level decay channels, accomplish the theoretical support of all project works by the analysis and processing of the experimental data.

The group headed by V.V. Lomonosov, Cand. Sci. (Phys-Math) (RRC "Kurchatov Institute") will carry out the experimental and theoretical developments of the methods for studying the chemical environment properties using the Thorium-229 isomer implanted into a solid or surface. It will carry out experimental investigations on the reduction of the optical background created by alpha-particles bremsstrahlung in the target material, study the isomer excitation by the tunable laser.

3.4.2. Planning.

III quarter of 1996. — Purchase of Thorium-229 and chemical reagents. Fabrication of the first targets on a metallic substratum and transparent matrix. Beginning of the measurements of target's background characteristic. Preparation of the experimental equipment for generation of surface plasmons. Beginning of the theoretical studies of the problem of gamma-laser in the optical range.

IV quarter of 1996. — Determination of the dynamics of the backgrounds of the targets on metallic substrata and transparent matrix. Development and tests of the methods for elimination of the backgrounds. Selection of laser emitters and bringing their characteristics to the desired level. Taking the atomic spectroscopy characteristics of the chemical Thorium compounds.

I and II quarters of 1997. — Experimental studies of excitation of the Thorium-229 isomeric state by surface plasmons. Determination of the decay channels, isomeric lifetime and energy.

II and III quarters of 1997. — Experimental studies of excitation of the Thorium-229 isomeric state by laser radiation. Pumping of the isomeric nucleus system.

IV quarter of 1997. — Theoretical and experimental studies of the Moessbouer effect in the optical range in the isomeric transition in Th-229.

I and II quarters of 1998. — Studies of the coherent gamma-radiation of isomeric Th-229 nuclei in the optical range. Development of the method for determination of the chemical environment properties by the photon spectrum of Th-229m decay through the electron bridge.

3.4.3. Equipment.

The teams have all the equipment required for the project accomplishment. A part of the funds will be spent for purchasing superpure chemical reagents and Thorium-229 for target manufacturing.

4.1. The total title of the project.

Investigation of the channels of excitation and decay of the anomalously low-energy isomeric level 3/2+ (3.5±1.0 eV) in the Th-229 nucleus.

4.2. Abstract.

The present project is aimed at investigation both of the superanomaly of the low-energy isomeric level 3/2+ (3.5±1.0 eV) in the Th-229 nucleus, and at studying a number of fundamental physical processes closely associated with the existence of this level. While working on the project the properties of the nuclear transition will be determined. In addition the isomer decay channels: the direct nuclear radiation in the optical range and the "electron bridge" i.e. third-order process will be studied. The measurements will be made at a large signal/noise ratio. For this purpose it is supposed to excite a great number of Th-229 nuclei for the isomeric state by both the laser radiation and surface plasmons via the inverse electron bridge (IEB) mechanism. The IEB process, where the atomic shell serves as a some kind of the electron bridge between the incident radiation and the nucleus, provides the resonant excitation of nuclei even in the presence of detuning between the photon energies (surface plasmons) and the nuclear transition energy. Upon the refinement of the isomer energy an accurate tuning of the radiation to the nuclear transition energy will become possible either via the system of atomic levels, or in a direct way. This will ensure a high-effective excitation or "pumping" of isomers and creation of the inverse population in the isomeric nucleus system. It is supposed to fabricate special targets on the basis of the optically transparent matrices for studying the coherent nuclear radiation and development of the laser on the nuclear transition to Th-229 in the optical range. Th-229m can be used as a supersensitive probe for studying the properties of surfaces and solids. In the process of the electron bridge decay the isomeric nuclei implanted into various samples will emit photons having the spectrum sensitive to the chemical environment and fine effects in the electron-nucleus and photon-nucleus interaction.

All the results to be obtained will be presented as publications in scientific journals and submitted at the international conferences.