Program: Capacitati / Modul III – Cooperare bilaterala/multilaterala

Contract Nr. 410/05.05.2010
Proiect: *"Studiul cristalelor de bromura de lantan ca nou material scintilator pentru aplicatii si cercetare stiintifica"*
Acronim proiect: NSLaB

Tara partenera: Turcia
Perioada de colaborare: 2010 – 2011

Institutii participante:
Romania: Intitutul National pentru Fizica si Inginerie Nucleara "Horia Hulubei", Bucuresti
Turcia: Universitatea din Istanbul

Prezentare generala a proiectului de colaborare

Scopul principal al acestui proiect este acela de a studia noii detectori de radiatii gama aparuti in ultimii ani, bazati pe materialul scintilator $LaBr_3$:Ce (bromura de lantan dopata cu ceriu). Acest material scintilator are o serie de proprietati care il fac extrem de interesant pentru spectroscopia gama si multe aplicatii bazate pe aceasta:

- rezolutie energetica mult mai buna decat alti scintilatori: tipic, 2-3% la 660 keV;
- rezolutie temporala foarte buna: 100 – 300 ps, depinzand de marimea cristalului;
- output luminos mare: peste 63 de fotoni/keV;
- timp de dezintegrare rapid: cca 16 ns (si fara nicio componenta de timp lenta).

Caracteristicile temporale si de rezolutie energetica il recomanda pentru cercetarile bazate pe spectroscopia gama unde se urmareste indeosebi masurarea timpilor de viata in domeniul de la picosecunda la nanonsecunda. Aici insa este necesar sa se studieze in amanunt semnalul de raspuns la radiatii gama de diferite energii, pentru cristalele de $LaBr_3$ disponibile comercial, si mai mult, sa se imagineze metode prin care sa se poata folosi un mare numar de module de acest tip in experimente tipice de spectroscopie nucleara (pentru cresterea eficientei de detectie).

In al doilea rand, buna rezolutie energetica, cuplata cu o eficienta de detectie rezonabila, fac ca aceste materiale scintilatoare sa devina interesante pentru multe aplicatii.

Atat in Institutul de la Bucuresti (IFIN-HH) cat si la Universitatea din Istanbul exista grupuri de cercetatori cu experienta deosebita in spectroscopia radiatiilor gama, motiv pentru care a fost propusa colaborarea in acest domeniu, cu scopul de a dezvolta aplicatii originale ale acestui tip nou de detectori atat in cercetarea stiintifica de baza, cat si in domenii aplicative ale acesteia.

Colaborarea are ca obiective:

- studiul raspunsului detectorilor de $LaBr_3$:Ce intr-un domeniu larg de energii ale radiatiilor gama, atat cu surse cat si in reactii produse de fascicule accelerate;
- studiul coincidentelor rapide gama-gama utilizand detectori de $LaBr_3$:Ce, eventual cuplati cu detectori de Germaniu hiperpur de inalta rezolutie energetica. Evaluarea posibilitatii de a utiliza acesti detectori rapizi pentru implementarea metodei electronice directe de masurat timpi de viata ai starilor excitate nucleare, in reactii nucleare induse de fascicule accelerate;
- investigarea posibilitatii de a utiliza raspunsul rapid al acestor detectori pentru a obtine informatii asupra pozitiei spatiale a surselor de radiatii, precum si alte tipuri de aplicatii.

Pe parcursul colaborarii sunt planificate multe serii de masuratori in comun, indeosebi in cadrul Departamentului de Fizica Nucleara al IFIN-HH, unde, in perioada mentionata, se afla in dezvoltare un dispozitiv multi-detector de masura a radiatiilor gama bazat pe detectori de Ge hiperpur si dtectori scintilatori $LaBr_3$:Ce.

Directori proiect:
pentru IFIN-HH: CS1 Dr. D. Bucurescu
pentru Universitatea Istanbul: Prof. Dr. M. Bostan

RAPORT FINAL DE ACTIVITATE

pentru proiectul de colaborare bilaterala Romania -Turcia:

„Studiul cristalelor de bromura de lantan ca un nou material scintilator pentru aplicatii si cercetare stiintifica"

Scopul principal al proiectului a fost studierea raspunsului noilor detectori de radiatii gama bazati pe $LaBr_3$:Ce. Dezvoltarea de noi materiale este o tendinta curenta in tehnologiile de detectie. In primii ani ai sec. 21, s-au descoperit doi noi scintilatori bazati pe "halide anhidre", $LaCl_3$:Ce si $LaBr_3$:Ce. Detectia radiatiilor electromagnetice ionizante (radiatii gama) este, mai intai de toate, o provocare pentru domeniul stiintei materialelor, si noul cristal de bromura de lantan dopat cu ceriu a aparut ca un nou material scintilator care umple distanta dintre scintilatorii conventionali de rezolutie energetica relativ joasa precum NaI(Tl), si detectorii de Germaniu hiperpur (HPGe) de inalta rezolutie, dar raciti criogenic sau cu azot lichid. Performantele superioare ale cristalelor $LaBr_3$:Ce pe intreg domeniul de energie al radiatiilor gama sunt reflectate intr-o inalte selectivitate si eficienta de detectie buna, mai buna decat cea a cristalelor de NaI(Tl). Un detector cilindric de $LaBr_3$:Ce de dimensiuni 2" X 2", de exemplu, are o rezolutie energetica de ~3% la 662 keV, producere de lumina de circa $61*10^3$ fotoni/MeV, si are excelente proprietati temporale: timp de dezexcitare de cca 16 ns si fara o componenta lenta intensa. Toate aceste proprietati sunt superioare materialelor scintilatoare folosite in mod curent, precum NaI(Tl). Recent, au aparut pe piata cristale din ce in ce mai mari, si este interesant de investigat daca aceste proprietati atractive sunt pastrate de cristalele produse prin procese comerciale. Aceste proprietati fac scintilatorii de $LaBr_3$ extrem de atractivi pentru toate domeniile care se bazeaza pe spectroscopia gama, de la cercetarile de structura nucleara la diferite aplicatii. Colaborarea prezenta s-a axat pe potentialul atat stiintific cat si aplicativ al acestor detectori.

Intr-o prima faza a proiectului, s-au testat cu surse radioactive gama calibrate de ^{152}Eu, ^{137}Cs si ^{60}Co, cristale de $LaBr_3$:Ce de diferite forme si dimensiuni – cristale cilindrice cu diametrele de 2" si 1.5", si cristale conice cu diametrul de 1.5". Selectarea surselor a fost facuta astfel incat sa se asigure masuratori omogene pe un domeniu energetic mare, pana la 1.4 MeV. Pentru toate cristalele, raspunsul detectorului a fost linearizat, si s-au masurat parametrii de raspuns – cheie, precum eficienta si rezolutia in energie si timp.

Pentru a utiliza detectorii de $LaBr_3$:Ce in experimente de spectroscopie nucleara, s-a dezvoltat si testat in colaborare un dispozitiv de masuratori experimentale, la Departamentul de Fizica Nucleara al IFIN-HH, Bucuresti. Dispozitivul include doua tipuri de detectori gama, $LaBr_3$:Ce si HPGe, pentru a optimiza informatia obtinuta – utilizand rezolutia energetica superioara a HPGe (pentru a obtine date asupra energiei si spinului nivelelor nucleare excitate prin intermediul radiatiilor gama ce dezexcita aceste nivele), si proprietatile temporale superioare ale $LaBr_3$:Ce (pentru a masura timpul de viata al unora din aceste nivele nucleare). Pentru a construi coincidente gama-gama intre toti acesti detectori, a fost configurata o schema electronica dedicata, si a fost folosita intr-un experiment de testare. Testul in fascicul a folosit reactia ^{197}Au + α; o tinta de Au cu grosimea de 9.65 mg/cm^2 a fost bombardata cu un fascicul de particule α cu energia de 24 MeV si intensitatea de 10 nA. Semnalele de timp ale detectorilor au fost prelucrate utilizand module de elctronica nucleara de tipul CFD (Discriminator la Fractiune Constanta). Utilizarea acestui tip de module induce o variatie cu energia gama a semnalului temporal la iesire, asa incat apar deplasari intre semnalele de timp ale diferitilor detectori rapizi ($LaBr_3$:Ce). Pentru a trata aceasta sursa de eroare, a fost dezvoltata o metoda pentru a corecta dependenta de energie printr-o functie polinomiala (determinata la prelucrarea off-line a datelor), si pentru a alinia in timp toti detectorii rapizi. Aceasta metoda prin care se construiesc coincidente gama-gama intre un mare numar de detectori $LaBr_3$:Ce si HPGe este noua, si prima de acest gen propusa pentru masuratori electronice rapide in fascicul folosind $LaBr_3$:Ce. In experimentul de test s-au re-masurat valori ale unor timpi de viata cunoscuti, si s-a demonstrat ca metoda este foarte precisa intr-un domeniu de la zeci de picosecunde la cateva nanosecunde.

Noua metoda a fost utilizata intr-un alt experiment efectuat la acceleratorul Tandem al IFIN-HH, pentru am masura valori de timpi de viata ale unor stari in nuclee exotice. S-a folosit o tinta de ^{18}O obtinuta prin incalzirea unei foite de Ta de grosime 50 mg/cm^2 in atmosfera de oxigen imbogatit izotopic pentru a obtine Ta_2O_5 cu o grosime echivalenta pentru ^{18}O de 1.6 mg/cm^2 pe ambele fete ale foitei de Ta. S-a utilizat un fascicul de ^{18}O de 36 MeV si o intensitate medie pe tinta de 20 pnA. Dispozitivul experimental a inclus 8 detectori HPGe si 7 cristale de $LaBr_3$:Ce de diverse geometrii: 3 cilindrici de 2”×2”, 2 cilindrici de 1.5”×1.5” si 2 conici de 1.5”x1”. Detectorii HPGe au fost plasati la ± 37 grade (sase dintre ei) fata de directia fasciculului, iar cei de $LaBr_3$:Ce la ± 45 grade. Datele obtinute au fost achizitionate in modul eveniment-cu-eveniment, cu o conditie de trigger cerand existenta unei coincidente intre (i) minim 3 HPGe, SAU (ii) minim 2 $LaBr_3$:Ce. S-au inregistrat un numar de 10^9 coincidente in $LaBr_3$:Ce, aceste evenimente fiind prelucrate off-line si sortate in matrici de coincidenta bidimensionale gama-gama si tridimensionale gama-gama-gama. Matricile au fost analizate cu pachetul de programe GASPware. Principalele nuclee produse in reactii au fost ^{33,34}P, ^{33}S si ^{30}S in canalele ce implicau evaporarea de 1 proton si 2 neutroni (1p2n), 1p1n, 3n, si respectiv, 1p1n1α. In afara informatiei asupra schemelor de nivele ale nucleelor produse, s-a determinat pentru prima data rata de tranzitie electromagnetica de la starea yrast $J^{\pi}=4^{-}$ de la 2305 MeV in nucleul ^{34}P din timpul de injumatatire masurat, de 2 ns, al acestei stari. Acest rezultat, impreuna cu raportul de amestec M2/E3, va permite o comparatie detaliata cu calcule de model in paturi cu configuratii intr-un spatiu de valenta limitat, care include excitatii in paturile protonice si neutronice $f_{7/2}$.

Pe parte aplicativa, a fost dezvoltata o noua metoda de masurare in-situ a umiditatii in structuri de constructie (pereti, fundatii, materiale de constructie, etc.). Masurarea nedistructiva a umiditatii in materialele de construtie poroase poate fi foarte importanta, in special in cazul restaurarii cladirilor si monumentelor istorice, deoarece da o indicatie clara asupra starii exacte a structurii si a punctelor unde e nevoie de intervenit.

Metoda utilizeaza detectorii recent dezvoltati de $LaBr_3$:Ce pentru a masura retro-imprastierea Compton a radiatiilor gama de la o sursa colimata, pe materialul de constructie investigat. Flexibilitatea metodei este asigurata de portabilitatea echipamentului si de faptul ca atat detectorul cat si sursa de radiatii pot fi plasate de aceiasi parte a obiectului masurat. Dispozitivul experimental a fost testat utilizand imprastierea radiatiei de 48.9 keV a unei surse de ^{241}Am pe un material de constructie comun (caramida rosie din argila arsa). S-a demonstrat ca intensitatea radiatiei gama imprastiate variaza cu umiditatea si ca masurand intensitatea picului Compton se poate deduce foarte usor gradul de umiditate.

In concluzie, s-a dezvoltat un nou dispozitiv experimental de spectroscopie gama ce foloseste un ansamblu mixt de detectori de bromura de lantan si de germaniu de inalta puritate, care s-a dovedit util in masurarea timpilor de viata nucleari prin metoda electronica in domeniul de la zeci de picosecunde la cateva nanosecunde, in masuratori in fascicul. Dispozitivul a fost testat atat cu surse radioactive cat si in experimente in fascicul. De asemenea, cautand diverse aplicatii ale detectorilor de bromura de lantan, a fost propusa o metoda de masurare nedistructiva a umiditatii in materoale de constructie poroase, care s-a dovedit utila si precisa.

Rezultatele obtinute in cadrul acestui proiect vor fi folosite si dezvoltate in cadrul unor colaborari stiintifice interantionale mai mari, in care ambii parteneri prezenti sunt implicati. Pe baza rezultatelor si expertizei obtinute in timpul ultimilor doi ani, sunt asteptate noi rezultate din experimentele viitoare. Acestea vor constitui, de asemenea, o buna baza pentru activitatile stiintifice care implica noi doctoranzi si masteranzi, contribuind in acest mod la imbunatatirea calitatii resurselor umane ale ambilor parteneri.

Director Proiect pentru IFIN-HH,
Dr. Dorel Bucurescu

Director Proiect pentru Universitatea din Istanbul,
Prof. Dr. Melih Bostan

Program: Capacities / Module III – Bilateral/multilateral cooperation

Contract No. 410/05.05.2010
Project: *"Study of the lanthanum bromide crystals as a new scintillator material for applications and scientific research"*
Project acronym: NSLaB

Partner country: Turkey
Collaboration period: 2010 – 2011

Participating Institutions :
Romania: "Horia Hulubei" National Institute for Physics and Nuclear Engineering, Bucharest
Turkey: University of Istanbul

General presentation of the collaboration project

The main goal of this project is to study the new gamma radiation detectors that appeared during the recent years, based on the scintillator material $LaBr_3$:Ce (lanthanum bromide doped with cerium). This scintillator material has some properties that make it extremely interesting for the gamma ray spectroscopy and applications based on it:

- much better energy resolution than that of the other scintillators: typically 2-3% at 660 keV;
- very good temporal resolution: 100 – 300 ps, depending on the size of the crystal;
- large light output: typically more than 63 photons/keV;
- fast decay time: about 16 ns (without a slow time component).

The time and energy resolution characteristics recommend this material for researches based on gamma ray spectroscopy where of major interest is the measurement of nuclear level lifetimes in the picosecond to nanosecond range. Nevertheless, for such a purpose, it is necessary to study in detail the output signals of these detectors for gamma-rays of different energies, for the $LaBr_3$ crystals commercially available, and, moreover, to imagine methods that allow the use of a large number of such modules in typical nuclear spectroscopy experiments (to increase the detection efficiency).

Second, the good energy resolution, together with a reasonable detection efficiency, make that this detecting material becomes interesting for many applications as well.

Both in the Bucharest Institute (IFIN-HH) and in the University of Istanbul there are groups of researchers with an outstanding experience in the spectroscopy of the gamma radiations, and for this reason a collaboration in this domain has been proposed, with the purpose of developing original applications of this new type of detectors, both for the basic scientific research and applications of this.

The objectives of this collaborations are:

- study of the response of $LaBr_3$:Ce detectors in a wide range of energies of the gamma radiations, both with radioactive sources and in nuclear reactions induced by accelerated ion beams;
- study of the fast gamma-gamma coincidences of $LaBr_3$:Ce detectors, eventually coupled with hyperpure Germanium detectors of high energy resolution. The assessment of the possibility to utilize these fast detectors in order to implement the direct electronic method of measuring lifetimes of excited nuclear states, in nuclear reactions induced by accelerated ion beams;
- investigation of the possibility of using the fast response of these detectors in order to get information on the space position of the radiation sources, as well as for other types of applications.

During this collaboration, many common measurements have been planned, especially at the Nuclear Physics Department of IFIN-HH, where, during the time of this collaboration a multi-detector system for the gamma radiation measurements, based on both hyperpure Ge and $LaBr_3$:Ce detectors, is under development.

Project directors:
for IFIN-HH: Senior researcher Dr. D. Bucurescu
for University of Istanbul: Prof. Dr. M. Bostan

FINAL ACTIVITY REPORT

for the billateral Romania -Turkey collaboration project:

„Study of the lanthanum bromide crystals as
a new scintillator material for applications and scientific research"

The primary goal of the project was to study the response of the novel $LaBr_3$:Ce gamma radiation detectors. The development of new materials is the current trend in detection technologies. During the first years of the 21^{st} century, two new „anhydrous halide" scintillators were discovered, $LaCl_3$:Ce and $LaBr_3$:Ce. The detection of ionising electromagnetic radiations (gamma rays) is first of all a challenge for the material science field, and the new crystal, $LaBr_3$:Ce has emerged as a scintillation material closing the gap between the relatively low resolution conventional scintillators like NaI(Tl) and high resolution cryogenically cooled hyper-pure Germanium detectors (HPGe). The superior performance of $LaBr_3$:Ce crystals over the whole range of gamma-ray energy is reflected in their high selectivity and good detection efficiency, better than that of NaI(Tl) crystal. A $LaBr_3$:Ce cylindrical 2" X 2" detector, e.g., has an energy resolution of ~3% at 662 keV, light yield of $61*10^3$ photons/MeV, and excellent time properties: about 16 ns de-excitation time without an intense slow component. All these properties are superior to the scintillator materials that are currently used on a large scale, like NaI(Tl). Recently, bigger crystals have become available, and it is interesting to investigate whether these attractive properties are retained in crystals produced with commercially scalable processes. These properties make the $LaBr_3$ scintillators extremely attractive for all domains involving gamma spectroscopy, from nuclear structure research to various applications. Our collaboration focussed on both the scientific and applicative potential of these detectors.

In a first phase of the project, $LaBr_3$:Ce crystals of various shapes and dimensions - cylindrical crystals of 2" diameter, cylindrical crystals of 1.5" diameter and conical crystals of 1.5" diameter - were tested using standard calibrated gamma radioactive sources of ^{152}Eu, ^{137}Cs and ^{60}Co. The selection of the sources was made in order to insure homogeneous measurements on a broad gamma energy range of up to 1.4 MeV. For all the crystals, the detector response was linearized, and key response parameters like efficiency, energy and time resolutions were measured.

In order to use the $LaBr_3$:Ce detectors in nuclear spectroscopy experiments, an experimental set-up was developed and tested, in collaboration, at the Nuclear Physics Department of IFIN-HH, Bucharest. The set-up includes two types of gamma detectors, $LaBr_3$:Ce and HPGe, in order to optimize the obtained information - it makes use of superior energy resolution of HPGe (in order to get data about the energy and spin of nuclear excited levels and the gamma radiations that de-excite these levels) and of the superior timing properties of $LaBr_3$:Ce (in order to obtain the value of the lifetime for some nuclear levels). In order to build gamma-gamma coincidences between the detectors, a dedicated electronic scheme was configured and used in an in-beam test. The in-beam test used the ^{197}Au + α reaction; the Au target of 9.65 mg/cm^2 thickness was bombarded with an α particles beam of 24 MeV energy and 10 nA intensity. The time signals of the detectors were processed using nuclear electronic modules of CFD type (Constant Fraction Discrimination). The use of this type of modules induces a variation of the output signal with the gamma energy, so that shifts between the time signals of different $LaBr_3$:Ce appear. In order to handle this error source, a method was developed to correct the energy dependence by an off-line determined polinomial function, and align in time all fast detectors. This method that builds gamma-gamma coincidences between a large number of $LaBr_3$:Ce and HPGe detectors is new and the first one proposed for in-beam fast timing lifetime measurements with $LaBr_3$:Ce. In the test experiment lifetime values that were already known were re-measured and the results proved the method to be very accurate in the range of tens of picoseconds up to nanoseconds.

The new method was used in another experiment made at the Tandem accelerator of IFIN-HH in order to measure lifetime values for nuclear states in exotic nuclei. A ^{18}O target was made by heating a Ta foil of 50 mg/cm^2 thickness in oxigen-enriched atmosphere in order to obtain Ta_2O_5, of estimated total equivalent ^{18}O thickness of 1.6 mg/cm^2 on both sides of the Ta foil. A ^{18}O beam of 36 MeV and mean intensity on target of 20 pnA was used. The experimental set-up included 8 HPGe detectors of 50% relative detection efficiency and 7 $LaBr_3$:Ce scintillation detectors with crystals of various geometries: 3 cylindrical of 2”×2”, 2 cylindrical of 1.5”×1.5” and 2 conical of 1.5”x1”. The HPGe detectors were placed at ± 37 degrees (six of them), respectively ± 90 degrees (two of them) from the beam direction, while the $LaBr_3$:Ce detectors were placed at ± 45 degrees from the beam direction. The data obtained in the experiment were acquired in the event-by-event mode by putting a trigger condition that asked for the existence of gamma coincidence between: (i) minimum 3 HPGe OR (ii) minimum 2 $LaBr_3$:Ce. A number of 10^9 gamma coincidences in $LaBr_3$:Ce was recorded, these events being then off-line processed, sorted in form of bi-dimensional gama-gama and tri-dimensional gama-gama-gama coincidence matrices, and analysed using the GASPware software package. The main nuclei that were produced in the reaction were ^{33,34}P, ^{33}S and ^{30}S in the channels involving evaporation of 1 proton and 2 neutrons (1p2n), 1p1n, 3n, and 1p1n1α, respectively. Besides information about the level schemes of the produced nuclei, the electromagnetic transition rate from the yrast $J^{\pi}=4^{-}$ state at 2305 keV in ^{34}P was determined for the first time from its measured half-life of 2 ns. This result, together with the M2/E3 mixing ratio will allow a detailed comparison with limited valence-space configuration mixing shell model calculations that include excitations in the proton and neutron $f_{7/2}$ shells.

On the applicative side, a new in-situ non-destructive method for measuring moisture in construction structures (walls, foundations, building materials, etc) was developed. The non-destructive measurement of moisture in porous construction materials can be very important, especially in the case of restoration of historical buildings and monuments, because it gives a clear indication on the exact state of the structure and on the points where intervention is needed.

The method involves the recently developed $LaBr_3$:Ce scintillation detectors, used to measure the Compton backscattering of the gamma radiations from a collimated source from the construction material under test. The flexibility of the method is insured by the equipment portability and by the fact that both the detector and the source can be placed on the same side of the measured object. The experimental set-up was tested using the scattering of the 48.9 keV radiation from a ^{241}Am source, on a common construction material (red fired-clay bricks). It was demonstrated that the intensity of the scattered gamma radiation varies with the humidity and that by measuring the intensity of the Compton peak, the moisture degree can be very easily deduced.

In conclusion, a new gamma-ray spectroscopy experimental set-up that uses a mixed array of both lantanum bromide detectors and high-purity germanium detectors was developed and proved useful for measuring nuclear lifetimes with the electronic method in the range of tens of picoseconds up to few nanoseconds, in in-beam experiments. The set-up was tested both with radioactive sources and in an in-beam experiment. Also, looking for applications of lantanum bromide detectors, a method for non-destructive measurements of moisture in porous construction materials was proposed, which was found to be both handy and accurate.

The results that were obtained in this project will be used and developed in the frame of larger international scientific collaborations in which both partners are involved. Based on the achievements and expertise obtained during the last two years, we expect many new results from future experiments. These will also constitute a good basis for the scientific activities involving new PhD and Msc students, helping in this way to improve the quality of the human resources of both partners.

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