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The ESQUIRE project: Quantum Dots as scintillation detectors

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Summary. — The primary goal of ESQUIRE (Experiment with Scintillating Quantum dots for Ionizing Radiation Events) is the development of a new family of scintillation detectors based on scintillating nanocrystals coupled to high-quantum-efficiency solid-state detectors. These detectors will be designed for the search of neutrinoless double-beta decay ($0\nu\beta\beta$), therefore an excellent energy resolution in the region of interest for the study of $0\nu\beta\beta$ ($\sim$2% around 3 MeV) is mandatory. One of the main advantages in this approach is the easy mass scalability, which makes ESQUIRE a competitive option for next-generation experiments. During the discussion the project goal will be presented, alongside the first optical characterization of QD samples.

1. – Introduction

The neutrinoless double-beta decay ($0\nu\beta\beta$) is an hypothesized nuclear decay in which a nucleus (A, Z) decays into a member $(A, Z+2)$ of the same isobaric multiplet with the emission of only two electrons. The eventual discovery of this process would state whether neutrinos are Dirac or Majorana fermions and provide information about their absolute mass scale, while proving the lepton number violation. The experimental signature of this process would be a peak at the Q-value of the transition ($Q_{\beta\beta}$) in the energy sum spectrum of the two emitted electrons. The detectability of this signal is firstly limited by the rarity of the process (current limits on the half-life of this decay are $\geq 10^{25}$ y), forcing the detector to reach extremely low levels of spurious background counts and to operate large masses of candidate isotopes ($\geq$ ton detectors). In addition, $0\nu\beta\beta$ is a concurrent process with respect to the two-neutrino double-beta decay ($2\nu\beta\beta$), characterized by a continuous spectrum reaching the $Q_{\beta\beta}$. To make the $2\nu\beta\beta$ contribution to the background negligible at the present half-life sensitivities of many $0\nu\beta\beta$ experiments, detectors searching for the $0\nu\beta\beta$ have to reach a FWHM energy resolution of the order of $\sim$2% at an energy of $\sim$3 MeV, a value close to the $Q_{\beta\beta}$ of most of the $0\nu\beta\beta$ candidate isotopes of interest. Current generation experiments are mainly limited either by the resolution request or by the mass scalability, depending on the chosen technology [1]. The ESQUIRE project aims to prove a novel approach to the search of $0\nu\beta\beta$ potentially capable of overcome all these limitations simultaneously. ESQUIRE proposes the development of a detector for $0\nu\beta\beta$ search based on the use of high-performance solid-state detectors (SDDs, silicon drift detectors) to read the light emitted by scintillating nanocrystals containing the $0\nu\beta\beta$ candidate isotope. The main goal of the initial phase of the project is to prove the feasibility of a nanocrystal based scintillation detector, capable of reaching good energy resolutions up to MeV energies.

2. – Quantum dots

The main ESQUIRE feature is the application of a new category of materials as scintillation light emitters: the quantum dots (QDs). QDs are semiconducting nano-crystals with diameters ranging between 2 and 10 nanometers. The electrons are therefore confined in a space smaller than the exciton Bohr radius, leading to the quantization of the energy. Given the nature of this quantum confinement, the available energy levels depend on the size of the QD. As a consequence, a precise control of the nanocrystal size allows the selection of the emission wavelength of the QD, which falls in the visible range, given the nanometer scale radius of the nanoparticle. In addition, the characteristics of the energy level structure allows an efficient and quick recombination, resulting in the quick (on the ns scale) release of scintillation photons. At the current state of the art, the production of heterostructured nanocrystals has been consolidated (Stokes-shift engineered QDs, *e.g.*, CdSe/ZnS [2]), allowing to tune the energy separation between the absorption and emission spectra by separating light absorption and emission functions between two distinct parts of the QD. This feature is fundamental, since low self-absorbing samples are needed to scale towards higher densities and dimensions without compromising the intensity of light output. The combination of these features makes QDs a good candidate for the development of wide scintillation detectors, both efficient and quick in light emission and capable of stopping $\sim$MeV ionizing radiation. During the ESQUIRE project different compounds such as CdZnS:Mn and CuInS/ZnS will be produced and analyzed, trying to develop a new family of scintillation detectors. Nevertheless, QDs containing isotopes candidate to $0\nu\beta\beta$ are needed to ensure the applicability of this technology to the search for $0\nu\beta\beta$. Among the possible isotopes, ^{116}Cd, ^{82}Se and ^{100}Mo are particularly interesting because of their transition energy higher than 2615 keV (practically the end of the environmental gamma-ray background spectrum) and their good natural abundances of 10%. Among the QDs containing these isotopes, $CuInSe_2$ (CISe) is a very promising one because of the high Stokes shift [3].

3. – The silicon drift detectors

The SDDs are solid state detectors with high quantum efficiency ($\sim$ 80% in the 450–1000 nm λ region) and low electronic noise. In particular, the anode capacitance of these detectors is extremely low (tens of fF) and does not depend on the active area of these devices [4]. This advantage of SDDs, as compared to other Si-based photodetectors, allows scaling of the detector size to effectively cover larger scintillating crystals with small electronic noise. The high quantum efficiency, combined with the possibility of reading the light output of large scintillators, limits the statistical resolution component, since a large number of photoelectrons can be generated. Moreover, the absence of charge multiplication mechanism avoids the resolution spoiling due to the intrinsic spread in the multiplication gain. It has already been demonstrated, *e.g.*, for CsI(Tl) [5], that SDDs can be used as photodetectors for scintillators readout, leading to improvements in terms of compactness, low voltage biasing, energy resolution, and linearity.

4. – Project steps

The ESQUIRE project is actually in a research and development phase (R&D) started at the beginning of 2018, divided into two parallel aspects: the optical characterization and selection of the QDs samples and the development and characterization of the SDDs. The optical characterization of the samples will guide the selection of the best QD sample. On one side, optical absorption and photo-luminescence emission/excitation measurements will be performed over a wide temperature range (10–320 K), in order to evaluate the temperature dependence of both the quantum efficiency and the self-absorption of the samples. On the other hand X-ray excited radioluminescence (RL) will be carried out, to evaluate the emission efficiency of the samples under ionizing radiation. Through these optical measurements, the samples with the highest light yield and optimal optical properties will be selected. In addition, the light yield dependence on the QD concentration, host material, and sample sizes will be also investigated, in order to optimize the final sample. The best solution will then be used to build a prototype detector. To further characterize the detector module, a series of Monte Carlo simulations will be carried out to evaluate the efficiency of the QDs as particle absorbers. This procedure will help both the detector development and the comprehension of future measurements. On the photodetector size, the development of new SDDs array will be conducted in collaboration with FBK in Trento. The optimization will implement on the one hand the enhancement of the quantum efficiency and on the other hand the eventual deposition of a protection layer to enhance the SDDs robustness. Different tests with SDDs equipped with low-noise CMOS preamplifiers (CUBE) [6] applied to the readout of QDs scintillators will be carried out to define the possible optimization steps. During the detector development, a parallel study on the processing electronics will be also conducted by means of digital pulse processing (DPP) on the output signal of the SDD+CUBE ensemble. Once a performing processing algorithm will be detected, a new electronics based on the use of DPPs or on new designed readout ASIC will be developed.

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